2 **Response to reviewer 1**

3 We are grateful to Reviewer for his valuable comments and remarks.

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Review of ACP-2017-508

9 In this manuscript the authors made theoretical assessment of how mixing of cloudy air with 10 unsaturated clear air affects the evolution of cloud microphysical parameters in the mixed volume, 11 using a one dimensional diffusion-evaporation model. Entrainment and mixing in clouds have been known to be an important but not well understood problem in cloud physics for several decades even 12 13 though their effects on cloud microphysics critically affect further cloud development. In that sense, 14 this type of study is highly needed for advancement of our understanding of the problem. The authors 15 calculated droplet spectral evolution in a one dimensional horizontal column of 40 m length during the turbulent mixing of cloudy air and unsaturated clear air of different proportions. Mixing starts at 16 17 the interface between cloudy and clear air and proceeds gradually throughout the whole length by turbulent diffusion. Cloud droplet spectrum in the mixed volume changes due to droplet evaporation 18 19 until saturation is restored. All these processes are expected to occur during the entrainment and 20 mixing and with the model they employed, the authors seem to have calculated these processes well. 21 But the critical question is if indeed they occur in real clouds under the conditions provided in this 22 manuscript. If not, it would be inappropriate to give so much credit to the arguments the authors made in this manuscript. Nevertheless, I think that this manuscript is worth the publication if the 23 24 authors clearly specify the limitation and applicability of their results. English is definitely not up to 25 the standard of ACP publication and therefore requires great improvement. Some specific comments are followed. 26

27

28 Major comments

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© According to the model description, turbulent diffusion and evaporation of the droplets in the
mixed volume occur simultaneously. The authors call this process inhomogeneous mixing because
the degree of mixing is not uniform throughout the whole volume during the mixing. I am not sure if
inhomogeneous mixing is the right term for this process.

³⁴ ® we state that mixing is inhomogeneous in a mixing volume because different droplets in the
³⁵ volume experience different subsaturations and evaporate with different rates. We believe that this
³⁶ definition is the logical and widely accepted.

© But I will stick to this definition for this review. In a one dimensional column, mixing may
proceed only this way no matter what Damkohler number (Da) is. It seems that the model is capable
of simulating homogeneous mixing as a special case for very small Da. How about the case of
extremely inhomogeneous mixing, which is likely to occur when Da is very large?

42

(8) Yes, we agree that the diffusional-evaporation model can describe both homogeneous and inhomogeneous mixing. Effects of Damkohler number on mixing within the frame of diffusional – evaporation model is analyzed in detail in Parts 2 and especially, in Part 3 of the study (Pinsky et al., 2016a,b). In particular, In Part 3 several types of mixing (homogeneous, intermediate, inhomogeneous and extremely inhomogeneous) are distinguished. In the present study we use parameters of mixing volume, turbulent diffusion and droplet size distribution, which as we suppose, are typical of **real** clouds. These parameters correspond to the values of Da of several hundred.

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© In a 3-D space overturning of turbulent eddies during the mixing of cloudy air with clear air may create some portion of cloud volume remaining unaffected and some other portion of cloud volume losing all droplets due to complete evaporation. Is the model capable of simulating something similar to this 3-D reality when Da is very large? Or setting a different Da value just changes the speed of mixing and evaporation that occur in an in variably simultaneous fashion in the mixed volume? If so, this model does have intrinsic limitation.

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8 We do not describe formation of separate turbulent filaments. In the study we describe averaged effects of turbulent mixing by modeling of turbulent diffusion, characterized by a typical value of turbulent diffusion coefficient. This is specially stressed in the paper. In principle, the model allows consideration of penetration and mixing of separate filaments by setting the specific initial conditions. However, we suppose that spatial-time distribution of cloudy and droplet free filaments is not well known and the investigation of the sensitivity of mixing to such distributions is out of scope of the present study.

65

© Another important point is that it takes nearly 5 min to arrive at the equilibrium state in the model
simulations. In reality the mixed volume of 40 m length would not remain as an adiabatic entity for
that long time as is pointed out by the authors themselves in the Discussion and Conclusion section.
Therefore, I am hesitant to give too much credit for the arguments based on the results obtained at the
equilibrium state.

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72 ® We agree with the reviewer. Moreover, it is the goal of this and some other our papers to show that

73 mixing in real clouds does not reach equilibrium state and that the scattering diagrams observed in

⁷⁴ situ are just snapshots of the transient mixing process. However, since the classic mixing diagrams

75 are plotted namely for equilibrium states, we investigate transition to such equilibrium assuming that

- 76 the mixing volume remains adiabatic (i.e. isolated) during the entire period of mixing.
- 77 This point is stressed in the revised paper.
- 78

© Narrow and wide DSDs are used as input to the model. It is shown that mixing and 79 80 evaporation actually result in increase of re in the mixed volume when DSD is wide. However, 81 assuming a wide DSD does not reflect the reality. In the argument of entrainment and mixing, we 82 start with an adiabatic cloud parcel and see how entrain and mixing of clear air would affect cloud 83 microphysics in this parcel. Importantly, droplet distributions in adiabatic parcels are intrinsically narrow. The wide DSD the authors used is therefore unrealistic. If a wide DSD is observed in a cloud 84 85 parcel, it would indicate that this parcel has already been through severe entrainment and mixing and/or coalescence process but certainly not a parcel that will start entrainment and mixing just now. 86

87

88 ® Indeed, pure diffusion growth leads to very narrow DSD. However, several other microphysical processes lead to DSD broadening. Mechanisms of DSD broadening in ascending 89 adiabatic volumes are considered in several studies (e.g., Khain et al. (2000), Pinky and Khain 90 (2002), Segal et al. (2004), Prabha et al. (2011)). These studies show that the DSD broadening is 91 92 caused by in-cloud nucleation of droplets within clouds as well as by collisions. It is shown that DSDs in adiabatic volumes can be wide and first raindrops or drizzle arise in non-diluted adiabatic 93 cloud parcels (Khain et al., 2013, Magaritz-Ronen et al., 2016). We use wide DSD in the form of 94 gamma distribution with parameters typically used in different bulk-parameterization models to 95 96 describe wide and narrow DSDs. We agree that mixing leads to additional DSD broadening. We also do not see any problem if the DSDs, which are used as initial DSDs in cloudy parcels were affected 97 by mixing during their previous history. It does not affect our analysis. The main point is that in these 98 99 parcels initially RH=100%.

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Corresponding comments are included into the revised text.

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- 102

© RH of 60% is the lowest in the model calculation of this manuscript. What about the mixing of
air with RH of~20%, which is a proper RH value for the air above the stratocumulus cloud top in the
subtropics? Mixing will be more likely to be extremely inhomogeneous when this very dry air is
entrained. Can this be simulated in the model?

107

108 [®] The model can work at any initial RH in the dry volume. At the same time very low RH leads 109 to total evaporation of droplets in the mixing volume. Cloud fraction should exceed 0.8 to get droplets in the equilibrium state at RH=20% (at LWC=1 g/kg). We believe that turbulence above the
stratocumulus cloud top is very weak so, cloud fraction should be large. At the lateral edges of warm
Cu a shell of humid air arises around cloud, so RH of the entrained air is higher than 20% (Gerber et
al. 2008).

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115

© The authors claim that mixing diagrams are not capable of distinguishing between mixing 116 117 types. Moreover, results are not distinguishably different for different RH values of entrained clear 118 air (Fig. 11). However, distinction between inhomogeneous and homogeneous mixing seems so 119 obvious in Fig. 9a. It is not so in Fig. 9b as the authors claim but here unrealistically wide DSD is used for the calculation and therefore giving too much credit is unjustifiable. Even in the traditional 120 121 mixing diagram that uses normalized concentration (N/N1) as in Fig. 11, distinction should be 122 obvious between inhomogeneous and homogeneous mixing and also among different RH values for homogeneous mixing. The authors do not put the lines for homogeneous mixing in Fig. 11. If they 123 124 do, I expect that the results would be distinctively different from those for inhomogeneous mixing.

125 Their argument is based only on the results for inhomogeneous mixing. However, RH 126 dependence of inhomogeneous mixing is not known to be significant anyway and has never been 127 discussed in mixing diagram analysis (e.g., Burnet and Brenguier, 2007).

128

129 ® The dispersion of points in situ measured mixing scattering diagrams is large. In the revised 130 paper we present as an example a scattering diagram taken from Burnet and Brenguier (2007) with 131 overloaded curves in Fig.9a (narrow DSD). Solid lines correspond to inhomogeneous mixing, while 132 dashed lines correspond to homogeneous mixing. One can see that the high scattering does not allow 133 to separate the mixing types.

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140 © Another thing to note is that in the traditional mixing diagram, y-axis is the cube of normalized mean volume radius (representing mean volume of the droplets), not the cube of normalized effective 141 radius as is used in this manuscript. With the obvious relationship of L = NV, where L is the liquid 142 143 water content, N is the number concentration and V is the mean volume of the droplets, L/L_a (i.e., normalized L, where L_a is the adiabatic value of L) = const lines can be drawn with rectangular 144 hyperbolic lines in the mixing diagram, making the diagram somewhat like a 3 D field of N, V and L 145 146 (Burnet and Brenguier, 2007). So I recommend the authors to use mean volume radius instead of 147 effective radius.

148

149 ® We agree with Reviewer that the mean volume radius could be used in the analysis instead of 150 effective radius. Note, however, that since effective radius in a wide range of conditions is only by 151 10% larger than the mean volume radius, the utilization of effective radius does not affect 152 significantly the results. Moreover, satellites measure specifically effective radius. Accordingly, 153 some authors (e.g. Freud et al., 2011) use effective radius for plotting the mixing diagrams.

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156

155 Minor comments

© L28: It is not right to say that droplet concentration remains unchanged when mixing is homogeneous. It does reduce because of simple dilution of cloud volume by clear air. The total number of droplets in the whole mixing volume remains unchanged but not the concentration, which is the number of droplets in a unit volume.

 © In several Figure captions, it is stated that p = 8288 mb. Shouldn't it be 828.8 mb? © Corrected © Figures 9, 10 and 11: How are re/re0 values obtained? Are they averages for the whole mixed volume at the time when equilibrium is achieved? How about N/N1? Explain clearly. © In these figures the mixing diagrams corresponding to the final equilibrium states are calculated. In the equilibrium state (about 300s) all quantities, including effective radius and droplet concentration become uniform throughout entire volume. © Inappropriate English expressions are found in many places in this manuscript. They need to be corrected. @ We have improved English whenever possible: 	.62 [®] Yes, we agree. The cor	responding sentence is corrected (line 28).
 Orrected • Figures 9, 10 and 11: How are re/re0 values obtained? Are they averages for the whole mixed volume at the time when equilibrium is achieved? How about N/N1? Explain clearly. • In these figures the mixing diagrams corresponding to the final equilibrium states are calculated. In the equilibrium state (about 300s) all quantities, including effective radius and droplet concentration become uniform throughout entire volume. • Inappropriate English expressions are found in many places in this manuscript. They need to be corrected. • We have improved English whenever possible. 	53 54 © In several Figure captio	ons, it is stated that p = 8288 mb. Shouldn't it be 828.8 mb?
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 be corrected. We have improved English whenever possible. 	© Inappropriate English	expressions are found in many places in this manuscript. They need to
(*) We have improved English whenever possible.	be corrected.	
	[®] We have improved Eng	glish whenever possible.

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194	We are grateful to Dr. Kumar (Reviewer 2) for valuable
195	comments and remarks.
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197	Theoretical analysis of mixing in liquid clouds. Part IV: DSD
198	evolution
100	and miving diagrams by M. Dinsky and A. Khain
199	and mixing diagrams by M. Pinsky and A. Kham
200	
201	
202	This study present analysis of mixing in cloudy and clear air. Evolution of DSD has
203	been analyzed using poly-disperse initial DSD and varying cloud fraction μ . A diffusion-
204	evaporation model was considered for the analysis. The main findings are
205	Mixing diagram has multi-parameter characteristics.
206	• In dry volume, mixing leads to a rapid increase in RH
207	• DSD shape changes based on initial DSD chosen.
208	• The critical cloud fraction μ_{cr} with respect to total droplet evaporation are same for
209	any mixing type.
210	• Mixing diagrams for homogeneous and in-homogeneous mixing for poly-disperse
211	DSD do not differ much.
212	
213	It was concluded that classical concept of mixing diagram is too crude to distinguish
214	the mixing type in observation data.
215	[®] Thank you for the clear summary of the study
216	
217	General comment: Overall, the manuscript is worth to publish after explaining
218	questions below.
219	
220	Major comments:
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222	1. Explain the reason for considering droplet concentration by averaging along X-
223	axis only why not in whole domain? Also, why vertical velocity was neglected? Since, the
224	analysis is done based on these assumptions, it is inappropriate to make strong general
225	statement about mixing diagrams.
226	
227	® The paper reconsiders the classical theory of mixing diagrams. In the classical
228	theory two volumes (cloudy and droplet free) mix with each other within a given

unmovable mixing volume (see review by Korolev et al., 2016). The 1D diffusionevaporation model is used in analysis. In this model all variables change along horizontal
x-axes (e.g. figs. 2-7). This model does not involve any spatial averaging. The vertical
velocity is also neglected in 1D model. Mixing diagrams are plotted for times when all
variables become uniform within the mixing volume, i.e when the equilibrium state is
reached.

We plot the mixing diagram applying widely used simplifications, namely: no vertical motions and no collisions. These assumptions allow to reveal better the microphysical effects of turbulent mixing. It is widely assumed that the mixing type is determined by the Damkohler number that the ratio of mixing time and drop relaxation time. Vertical velocity and collision rate are not included into this criterion.

We extend the classical theory in several important aspects concerning microphysical effects: a) we consider time dependent process of mixing and b) initial droplet size distributions are assumed polydisperse. These simplifications are clearly formulated in the paper.

We agree that averaged vertical velocity, as well as collisions, affect DSDs, but these changes in DSDs are not related to mixing, and are described by other microphysical equations.

2. In this analysis, collision and coalescence was not considered which also contribute in broadening of DSD. Authors should make comments on this issue.

8 The corresponding comments are included in the discussion section. We agree that many microphysical processes lead to the DSD broadening. The new feature that we stress in the study is that any mixing leads to DSD broadening (in contrast to conclusions of classical theory, considering monodisperse DSDs.) The corresponding comment is added into the conclusion section.

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3. The result shows that in dry volume large, droplets do not change their size significantly. This is not the case in general because during mixing, droplet size starts decreasing as soon as they enter in dry volume. Authors should provide the reason for it.

[®] According to equation of diffusion growth/evaporation the rate of droplet radii in subsaturation conditions decreases is inverse proportional to droplet radius. It means that if, say, 2
um radius droplet decreases twice during a certain time instance, the radius of 20 um droplet
will decrease by less than 0.1 um. It means that relative decrease in the sizes of large droplets is
much lower than that of small ones. The initial dry volume can faster saturate that hiders
further evaporation of larger droplets. Actually reduction in larger droplet sizes can be

267	insignificant. That is why we wrote that the size of large droplets remains approximately
268	unchanged.
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271	4. Traditional mixing diagrams should be plotted for normalized values of cube
272	radii vs. number concentration and then compare with mixing diagrams proposed in this
273	study.
274	
275	[®] Traditional (classical) mixing diagrams are plotted for monodisperse DSDs. In this
276	case the cloud fraction is equivalent to the normalized concentration. In the present study we
277	plotted scattering and mixing diagrams both as dependencies of normalized values of cube
278	radii on cloud fraction (figs. 8-10) and on number concentration (Fig 11)
279	
280	Minor comments:
281	1. All figure labels, legends should be bigger size to be visible enough.
282	[®] improved
283	
284	2. Some references related to recent numerical simulation of entrainment and
285	mixing should be added.
286	
287	^(B) References to recent studies (Bera et al., 2016a,b; Kumar et al., 2014; 2017; Khain
288	et al, 2017, Yum et al., 2016) are added.
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304	Theoretical analysis of mixing in liquid clouds. Part IV: DSD evolution
305	and mixing diagrams
306	
307	Mark Pinsky, and Alexander Khain
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309	Department of Atmospheric Sciences, The Hebrew University of Jerusalem, Israel
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315	Submitted to
316	Atmospheric Chemistry and Physics
317	May 2017
318	Revised September 2017
319	
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326 Abstract

327 Evolution of droplet size distribution (DSD) due to mixing between cloudy and dry volumes is investigated for different values of the cloud fraction and different initial DSD 328 shapes. The analysis is performed using a diffusion-evaporation model which describes time-329 dependent processes of turbulent diffusion and droplet evaporation within a mixing volume. 330 Time evolution of the DSD characteristics such as droplet concentration, LWC, mean volume 331 332 and the effective radii is analyzed. The mixing diagrams are plotted for the final mixing stages. It is shown that the difference between the mixing diagrams for homogeneous and 333 inhomogeneous mixing is insignificant and decreases with an increase in the DSD width. The 334 335 dependencies of normalized cube of the effective radius on the cloud fraction were compared with those on normalized droplet concentration and found to be quite different. In case the 336 normalized droplet concentration is used, mixing diagrams do not show any significant 337 dependence on relative humidity in the dry volume. 338

The main conclusion of the study is that traditional mixing diagrams cannot serve as areliable tool for analysis of mixing type.

341

342 **Keywords:** turbulent mixing, droplet evaporation, DSD evolution, mixing diagram

The effects of mixing of cloudy air with surrounding dry air on cloud microphysics are still 345 the focus of many studies (see overview by Devenish et al., 2012). Processes of mixing are 346 investigated in observations (Yum et al., 2015; Bera at al., 2016a,b), Large Eddy Simulations 347 (Andrejczuk et al., 2009; Khain et al., 2017), Direct Numerical Simulations (Kumar et al., 2014, 348 2017). Processes of mixing and their effects on droplet size distributions were recently 349 investigated in a set of theoretical studies (Yang et al., 2016; Korolev et al., 2016 (hereafter, 350 Pt1); Pinsky et al., 2016 a,b). The Pt1 presented analysis of conventional (classical) concept of 351 mixing and introduced the main parameters characterizing homogeneous and extremely 352 353 inhomogeneous mixing. At a monodisperse initial droplet size distribution (DSD), homogeneous mixing leads to a decrease in droplet size and droplet mass content, while the number of droplets 354 remains unchanged. Extremely inhomogeneous mixing is characterized by decreasing the 355 number of droplets due to full evaporation of some fraction of droplets penetrating the initially 356 dry air volume while the DSD shape in the cloud volume remains unchanged. As a result of 357 358 extremely inhomogeneous mixing, droplet concentration decreases while the mean volume radii 359 remain unchanged. At a polydisperse DSDs, the extreme homogeneous mixing is characterized by proportional changes in DSD for all droplet radii (Pt1). Since widely used mixing diagrams 360 361 describe the final equilibrium stage of mixing they do not contain information about changes in microphysical quantities in the course of mixing. 362

Pinsky et al. (2016a, hereafter Pt2) analyzed the time evolution of monodisperse and polydisperse DSD during homogeneous mixing. It was shown that result of mixing strongly depends on the shape of the initial DSD. At a wide DSD, evaporation of droplets (first of all, of the smallest ones) is not accompanied by a decrease in the effective radius. Moreover, this radius may even increase over time. This result indicates that the widely used criterion of separation of mixing types based on the behavior of the effective radius during mixing is not generally relevant and may be wrong in application to real clouds.

Pinsky et.al. (2016b, hereafter Pt3) introduced a diffusion-evaporation model which 370 371 describes evolution DSDs and all the microphysical variables due to two simultaneously occurring processes: turbulent diffusion and droplet evaporation. Mixing between two equal 372 volumes of subsaturated and cloudy air was analyzed, i.e. it was assumed that the cloud 373 volume fraction $\mu = 1/2$. The initial DSD in the cloudy volume was assumed monodisperse. 374 These simplified assumptions allowed to reduce the turbulent mixing equations to two-375 parametric ones. The first parameter is the Damkölher number, Da, which is the ratio of the 376 377 characteristic mixing time to the characteristic phase relaxation time. The second parameter is the potential evaporation parameter R characterizing the ratio between the amount of water 378 vapor needed to saturate the initially dry volume and the amount of available liquid water in 379 380 the cloudy volume.

Within the Da - R space, in addition to the two extreme mixing types defined in the 381 382 classical concept, two more mixing regimes were distinguished, namely, intermediate and inhomogeneous mixing. It was shown that any type of mixing leads to formation of a tail of 383 small droplets, i.e. to DSD broadening. It was also shown that the relative humidity in the 384 385 initially dry volume rapidly increases due to both water vapor diffusion and evaporation of penetrating droplets. As a result, the effective radii in the initially dry volume rapidly 386 approach the values typical of cloudy volume. At the same time, the liquid water content 387 (LWC) remains significantly lower than that in the cloudy volume during much longer time 388 than required for the effective droplet radius to grow. 389

In the present study (Pt4) we continue investigating the turbulent mixing between an initially dry volume and a cloudy volume. The focus of the study is investigation of DSD temporal evolution and analysis of the final equilibrium DSD. In comparison to Pt3, the problem analyzed in this study is more sophisticated in several aspects: • The dependences of different mixing characteristics on cloud volume fraction $0 \le \mu \le 1$ are analyzed. In this case the equations of turbulent mixing cannot be reduced to the twoparametric problem as it was done in Pt3.

397 • The initial DSDs in cloud volume are polydisperse. We use both narrow and wide initial DSD described by Gamma distributions with different sets of parameters. The DSD are 398 the same as those used in Pt2. Mechanisms of formation of wide DSDs in clouds including 399 DSDs in undiluted cloud cores were investigated in several studies [e.g., Khain et al., 2000; 400 Pinky and Khain, 2002; Segal et al., 2004; Prabha et al., 2011]. These studies show the DSD 401 broadening is caused by in-cloud nucleation of droplets within clouds as well as by collisions. 402 It was shown that DSDs in adiabatic volumes can be wide and first raindrops or drizzle drop 403 arise namely in non-diluted adiabatic cloud parcels [Khain et al., 2013; Magaritz-Ronen et al., 404 2016]. We use both narrow and wide DSDs in the form of Gamma distribution with typical 405 parameters used in different cloud resolving models. The DSDs that are used as initial ones in 406 cloudy volumes could be formed also under influence of mixing during their previous history. 407

408 It does not affect our analysis.

The equation for supersaturation, used in this study, is valid at low humidity in the
initially dry volume and is more general and compared with that used in Pt3, which makes the
DSD calculations more accurate.

At the same time, some simplifications used in Pt3 are retained in this study. The vertical movement of the entire mixing volume is neglected; collisions between droplets and droplet sedimentation are not allowed. Also, we consider a 1D diffusion-evaporation problem. We neglect the changes of temperature in the course of mixing, which is possibly a less significant simplification. All these simplifications allow to reveal the effects of turbulent mixing and evaporation on DSD evolution.

418

419 2. Formulation of the problem and model design

In this study, the process of mixing is investigated basing on the solution of 1D diffusionevaporation equation (see also Pt3). According to this equation, evaporation of droplets due to negative supersaturation in the mixing volume takes place simultaneously with turbulent mixing. Since droplets within the volume are under different negative supersaturation values until the final equilibrium is reached, the modeled mixing is inhomogeneous. The droplets can evaporate either partially or totally. The evaporation leads to a decrease in droplet sizes and in droplet concentration.

Like in Pt3, the process of turbulent diffusion is described by a 1D equation of turbulent diffusion. The equation does not describe formation of separate turbulent filaments. Instead, it describes averaged effects of turbulent mixing by modeling of turbulent diffusion, characterized by a typical value of turbulent diffusion coefficient K. The mixing is assumed to be driven by isotropic turbulence at scales within the inertial sub-range where Richardson's law is valid. In

this case, turbulent coefficient is evaluated as in Monin and Yaglom (1975).

433
$$K(L) = C\varepsilon^{1/3}L^{4/3}$$
 (1)

In Eq. (1) ε is the turbulent kinetic energy dissipation rate and C = 0.2 is a constant (Monin and Yaglom, 1975), Boffetta and Sokolov (2002). Eq. (1) means that we consider the effects of turbulent diffusion at scales much larger than the Kolmogorov microscale, i.e. the effects of molecular diffusion are neglected. In the simulations, we use $L = 40 \ m$ and $\varepsilon = 20 \ cm^2 s^{-3}$.

438

439 *Geometry of mixing and the initial conditions*

440 The conceptual scheme presenting mixing geometry and the initial conditions used in the441 following analysis are shown in Figure 1.

442

443 **Fig 1 here**

445 At t=0 the mixing volume of length L is divided into two volumes: the cloud volume of length μL (Fig.1, left) and the dry volume of length $(1-\mu)L$ (Fig.1, right), where $0 \le \mu \le 1$ 446 is the cloud volume fraction. The entire volume is assumed closed, i.e. adiabatic. At t = 0 the 447 cloud volume is assumed saturated, so the supersaturation $S_1 = 0$. This volume is also 448 characterized by the initial distribution of the square of the droplet radii $g_1(\sigma)$, where $\sigma = r^2$. 449 liquid water 450 The initial mixing ratio in the cloudy volume is equal to $q_{w1} = \frac{4\pi\rho_w}{3\rho_\sigma} \int_0^{\infty} \sigma^{3/2} g_1(\sigma) d\sigma$. The integral of $g_1(\sigma)$ over σ is equal to the initial droplet 451

452 concentration in the cloud volume $N_1 = \int_0^\infty g_1(\sigma) d\sigma$. The initial droplet concentration in the 453 dry volume is $N_2 = 0$, the initial negative supersaturation in this volume is $S_2 < 0$ and the 454 initial liquid water mixing ratio $q_{w2} = 0$. Therefore, the initial profiles of these quantities 455 along the *x*-axis are step functions:

456

457
$$N(x,0) = \begin{cases} N_1 & \text{if } 0 \le x < \mu L \\ 0 & \text{if } \mu L \le x < L \end{cases}$$
(2a)

458
$$S(x,0) = \begin{cases} 0 & \text{if } 0 \le x < \mu L \\ S_2 & \text{if } \mu L \le x < L \end{cases}$$
(2b)

459
$$q_w(x,0) = \begin{cases} q_{w1} & \text{if } 0 \le x < \mu L \\ 0 & \text{if } \mu L \le x < L \end{cases}$$
 (2c)

460

461 The initial profile of droplet concentration is shown in Fig. 1b. This is the simplest 462 inhomogeneous mixing scheme, wherein mixing takes place only in the x-direction, and the 463 vertical velocity is neglected.

464 Since the total volume is adiabatic, the fluxes of different quantities through the left and 465 right boundaries at any time instance are equal to zero, i.e.

467
$$\frac{\partial N(0,t)}{\partial x} = \frac{\partial N(L,t)}{\partial x} = 0; \quad \frac{\partial q_w(0,t)}{\partial x} = \frac{\partial q_w(L,t)}{\partial x} = 0; \quad \frac{\partial q_v(0,t)}{\partial x} = \frac{\partial q_v(L,t)}{\partial x} = 0 \quad (3)$$

468 where q_{v} is the water vapor mixing ratio.

To investigate of mixing process for different initial DSD, we assume that DSD in the cloudvolume can be represented by a Gamma distribution:

471
$$f(r,t=0) = \frac{N_0}{\Gamma(\alpha)\beta} \left(\frac{r}{\beta}\right)^{\alpha-1} \exp\left(-\frac{r}{\beta}\right)$$
 (4)

where N_0 is an intercept parameter, α is a shape parameter and β is a slope parameter of distribution. The DSD f(r) relates to distribution $g_1(\sigma)$ as $f(r) = 2rg_1(\sigma)$. We performed simulations with both initially wide and narrow DSDs. The width of DSD is determined by a set of parameters. The parameters of the initial Gamma distributions used in this study are presented in **Table 1**. Parameters of the distributions are chosen in such a way that the modal radii of DSD and the values of LWC are the same for both distributions. These distributions were used in Pt2 for analysis of homogeneous mixing.

479

480 Table 1 here

481

482

483 *Conservative quantity* $\Gamma(x,t)$

The supersaturation equation for an adiabatic immovable volume can be written in the

485 form
$$\frac{1}{S+1}\frac{dS}{dt} = -A_2\frac{dq_w}{dt}$$
, where S is supersaturation over water, and the coefficient

486 $A_2 = \frac{1}{q_v} + \frac{L_w^2}{c_p R_v T^2}$ is slightly dependent on temperature (Korolev and Mazin, 2003) (notations

487 of other variables are presented in Appendix). In our analysis we consider A_2 to be a

488 constant. As follows from the supersaturation equation, the quantity

490
$$\Gamma(x,t) = \ln[S(x,t)+1] + A_2 q_w(x,t)$$
 (5)

492 is a conservative quantity, i.e. it is invariant with respect to phase transitions. In Eq. (5), 493 |S(x,t)| can be comparable with unity by the order of magnitude. The conservative quantity 494 $\Gamma(x,t)$ obeys the following equation for turbulent diffusion

495

496
$$\frac{\partial \Gamma(x,t)}{\partial t} = K \frac{\partial^2 \Gamma(x,t)}{\partial x^2}$$
(6)

497

498 with the adiabatic (no flux) condition at the left and right boundaries $\frac{\partial \Gamma(0,t)}{\partial x} = \frac{\partial \Gamma(L,t)}{\partial x} = 0$

499 and the initial profile at
$$t = 0$$

500

501
$$\Gamma(x,0) = \begin{cases} A_2 q_{w1} & \text{if } 0 \le x < \mu L \\ \ln[S_2 + 1] & \text{if } \mu L \le x < L \end{cases}$$
(7)

502

From Eq. (7) it follows that $\Gamma(x,0)$ is positive in the cloud volume and negative in the initially dry volume. The mean value of function $\Gamma(x,0)$ can be written as follows:

506
$$\overline{\Gamma} = \frac{1}{L} \int_{0}^{L} \Gamma(x,0) dx = \frac{A_2 q_{w1}}{L} \int_{0}^{\mu L} dx + \frac{\ln[S_2 + 1]}{L} \int_{\mu L}^{L} dx = \mu A_2 q_{w1} + (1 - \mu) \ln[S_2 + 1] \quad (8)$$

507

508 $\overline{\Gamma}$ can be either positive or negative. In the latter case a complete evaporation of droplets in the 509 course of mixing takes place.

510 The solution of Eq. (6) with the initial condition (7) is (Polyanin et al., 2004):

$$\Gamma(x,t) = \sum_{n=0}^{\infty} a_n \exp\left(-\frac{Kn^2 \pi^2 t}{L^2}\right) \cos\left(\frac{n\pi x}{L}\right) =$$
511 $\mu A_2 q_{w1} + (1-\mu) \ln\left[S_2 + 1\right] -$
2 $\left(\ln\left[S_2 + 1\right] - A_2 q_{w1}\right) \sum_{n=1}^{\infty} \frac{\sin(n\pi\mu)}{n\pi} \exp\left(-\frac{Kn^2 \pi^2 t}{L^2}\right) \cos\left(\frac{n\pi x}{L}\right)$
(9)

512 One can see that function $\Gamma(x,t)$ depends on three independent parameters A_2q_{w1} , S_2 and μ . 513 This function does not depend on the shape of the initial DSD in the cloud volume. In the final 514 state when $t \to \infty$, $\Gamma(x,t)$ is :

515
$$\Gamma(t=\infty) = \mu A_2 q_{w1} + (1-\mu) \ln[S_2 + 1]$$
 (10)

516 Therefore, $\Gamma(t = \infty)$ depends on the cloud fraction and the initial values of liquid water 517 mixing ratio in the cloud volume and the relative humidity in initially dry volume.

The final equilibrium values of supersaturation $S(x,\infty)$ and liquid water mixing ratio 519 $q_w(x,\infty)$ can be calculated using Eq. (5). The case $\Gamma(t=\infty) > 0$ corresponds to the

520 equilibrium state with
$$S(x,\infty) = 0$$
 and $q_w(x,\infty) = \mu q_{w1} + (1-\mu) \frac{\ln[S_2+1]}{A_2}$. The case

521 $\Gamma(t=\infty) < 0$ corresponds to the equilibrium state with $q_w(x,\infty) = 0$ and 522 $S(x,\infty) = (1+S_2)^{1-\mu} \exp(\mu A_2 q_{w1}) - 1$. At given q_{w1} and S_2 , there is a critical value of the 523 cloud fraction μ_{cr} which separates these two possible final equilibrium states. This critical 524 value corresponds to $\Gamma(t=\infty) = 0$ and can be calculated from Eq. (10) as:

525

526
$$\mu_{cr} = \frac{\ln[S_2 + 1]}{\ln[S_2 + 1] - A_2 q_{w1}}$$
(11)

527

528 Another expression for μ_{cr} was formulated in Pt1.

529 The examples of spatial-temporal variations of function $\Gamma(x,t)$ for different cloud 530 fractions and initial RH=80% are shown in **Figure 2.**

532 Fig 2 here

533

Upper panels $\mu = 0.1$ correspond to the case of final total droplet evaporation and negative 534 final function Γ , whereas the middle and bottom rows $\mu = 0.5$ and $\mu = 0.9$ illustrate partial 535 evaporation cases when the total mixing volume reaches saturation. It is interesting that the 536 time required for the final equilibrium state to be reached practically does not depend on the 537 cloud fraction, being ~180 seconds for the illustrated cases. The cases $\mu = 0.1$ and $\mu = 0.9$ 538 demonstrate a strong non-symmetric spatial variability of $\Gamma(x)$ function during the first 50 539 At $\mu = 0.5$, a nearly full compensation between saturation deficit in the dry seconds. 540 volume and available liquid water in the cloud volume takes place if at the equilibrium state 541 $S(x,\infty) = q_w(x,\infty) = \Gamma(x,\infty) = 0$. However, the compensation at $\mu = 0.5$ is not full because of 542 the nonlinearity of Γ in Eq. (5). 543

544

545 *Diffusion-evaporation equation for DSD*

To formulate the diffusion-evaporation equation we use a simplified equation for droplet evaporation (Pruppacher and Klett, 1997), in which the curvature term and the chemical composition term are omitted

549
$$\frac{d\sigma}{dt} = \frac{2S}{F}$$
 (12)

550 where $F = \frac{\rho_w L_w^2}{k_a R_v T^2} + \frac{\rho_w R_v T}{e_w (T)D} = const$ (Notations of other variables are presented in Appendix.)

551 The solution of Eq. (12) is

552
$$\sigma(t) = \frac{2}{F} \int_{0}^{t} S(t') dt' + \sigma_0$$
(13)

Eq. (13) means that in the course of evaporation, distribution $g(\sigma)$ shifts to the left without changing its shape. The diffusion-evaporation equation for function $g(x,t,\sigma)$ can be written in the form

556

557
$$\frac{\partial g}{\partial t} = K \frac{\partial^2 g}{\partial x^2} + \frac{\partial}{\partial \sigma} \left(\frac{d\sigma}{dt} g \right)$$
(14)

558 Combining Eqs. (12) and (14) yields

559
$$\frac{\partial g(x,t,\sigma)}{\partial t} = K \frac{\partial^2 g(x,t,\sigma)}{\partial x^2} + \frac{2S}{F} \frac{\partial g(x,t,\sigma)}{\partial \sigma}$$
(15)

560

Eq. (15) is similar to the diffusion-evaporation equation for size distribution function used in Pt 3. The first term on the right hand side of Eq. (15) describes the effect of turbulent diffusion, while the second term describes the changes of size distribution due to droplet evaporation. To close this equation, one can use Eq. (5) written as

565

566
$$S(x,t) = \exp[\Gamma(x,t) - A_2 q_w(x,t)] - 1,$$
 (16)

567

and the equation for liquid water mixing ratio

569

570
$$q_w(x,t) = \frac{4\pi\rho_w}{3\rho_a} \int_0^\infty \sigma^{3/2} g(x,t,\sigma) d\sigma$$
 (17)

The equation system (15-17) for distribution $g(x,t,\sigma)$ should be solved under the following initial condition

573
$$g(x,0,\sigma) = \begin{cases} g_1(\sigma) & \text{if } 0 \le x < \mu L \\ 0 & \text{if } \mu L \le x < L \end{cases}$$
(18)

574 and using the Neumann boundary conditions

576
$$\frac{\partial g(0,t,\sigma)}{\partial x} = \frac{\partial g(L,t,\sigma)}{\partial x} = 0$$
(19)

These equations were solved numerically on a linear grid of droplet radii r_j being within the range 0-50 µm, where j = 1...50 are the bin numbers. The number of grid points along the x-axis was set equal to 81. In numerical calculations, the "evaporation term" in Eq. (15) was approximated as

582
$$\frac{2S}{F} \frac{\partial g(x,t,\sigma)}{\partial \sigma} \approx \frac{g\left(x,t,\sigma + \frac{2S}{F}\Delta t\right) - g\left(x,t,\sigma\right)}{\Delta t}.$$
 (20)

583

A shift and subsequent remapping of DSD using the method proposed by Kovetz and Olund's (1969) were implemented to solve Eq. (20) with the help of MATLAB solver PDEPE. After calculation of $g(x,t,\sigma_j)$ function, DSD $f(x,t,r_j)$ was calculated using the relationship $f(x,t,r_j) = 2r_i g(x,t,\sigma_j)$.

588

589 3. Spatial-temporal variations of DSD and of DSD parameters

590 Mixing may take a significant time. Cloud microphysical parameters measured in *in-situ* 591 observations correspond to different stages of this transient mixing process. During mixing, 592 DSDs and its parameters change substantially, which makes it reasonable to analyze these 593 time changes.

Figure 3 shows time evolution of initially narrow DSD in the centers of the cloudy volume and of the initially dry volume. The values of DSD in the initially cloudy volume decrease while there are no significant changes in the DSD shape. At $\mu = 0.7$, the droplet radius corresponding to the DSD maximum remains unchanged during mixing staying equal to 10 μm . At $\mu = 0.3$ the effect of droplet diffusion on DSD is stronger, and mixing leads not only to a decrease in the DSD values, but also to a decrease in the peak droplet radius in the cloudy volume. Both at $\mu = 0.3$ and $\mu = 0.7$, mixing leads to broadening of the initial DSD due to the appearance of the tail of small droplets.

In the center of the initially dry volume, the rate of the DSD growth depends on the value of the cloud fraction. At a low cloud fraction, DSD maximum remains substantially lower for the most period of mixing than that in the cloudy volume. At the same time, the radius corresponding to the DSD maximum increases reaching 80% of its maximum value already within the first 5 s. This is due to the fast increase in the relative humidity during mixing, so large droplets penetrating the initially dry volume do not decrease in size anyhow significantly determining the values of modal, mean volume and effective radii.

609

```
610 Figure 3 here
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611

At the initially wide DSD (**Figure 4**), the radii of the DSD maximum do not change. It means that at the initial RH= 80%, mixing and evaporation leads to a fast saturation of the initially dry volume, after which the peak radius remains unchanged.

615

616

```
617 Figure 4 here
```

618

619

It is interesting that at $\mu = 0.3$ in the initially dry volume, DSD reaches its maximum during the transition period (Fig.4, at t=80s), and then decreases toward the equilibrium state. This behavior is caused by the competition between the diffusion and droplet evaporation.

Figure 5 shows spatial dependences of droplet concentration, LWC and the effective radius within the mixing volume at different time instances at narrow initial DSD. At small

values of the cloud fraction, diffusion of water vapor and droplets, as well as droplet 625 evaporation lead to a fast decrease in droplet concentration and in LWC in the initially cloud 626 volume. The effective radius in this volume decreases by about 12% in the course of mixing. 627 It is natural that at large cloud fraction, droplet concentration and LWC in the initially cloudy 628 volume decrease slowly, while these quantities in the initially dry volume increase rapidly At 629 both small and large cloud fractions, the effective radius in the initially dry volume grows 630 rapidly during the mixing toward its values in the initially cloudy volumes, even if droplet 631 concentration and LWC remain much lower than in the cloud volume. 632

633

634 **Figure 5 here**

635

Figure 6 shows the spatial dependences of droplet concentration, LWC and the effectiveradius within the mixing volume at different time instances at wide initial DSD.

638

639

640 **Figure 6 here**

641

642

A specific feature of mixing at a wide DSD is the increase in the effective radius, so the ratio $\frac{r_e}{r_{e0}} > 1$. In the course of mixing, the effective radius maximum is reached in the initially dry volumes. This result can be attributed to the fact that in this volume smaller droplets fully evaporate, so the concentration of large droplets increases with respect to concentration of smaller droplets (Fig. 4, right column). Scattering diagrams plotted using *in-situ* observations often contain points or groups of points with $\frac{r_e}{r_{e0}} > 1$ (e.g., Burnet and Brenguier, 2007; Krueger et al., 2006, Gerber et al., 2008). In these observations, the effective radius was 650 measured within the cloud volume with maximum LWC (i.e. less diluted). The result obtained 651 in the present study shows that the behavior of $\frac{r_e}{r_{e0}}$ with time in the course of mixing may

652 depend of the DSD shape in the initially cloud volume.

We see that the transition to the final equilibrium state within the volume with the spatial 653 scale of 40 m is about 5 min (Fig. 8), which is a comparatively long period of time compared 654 655 to the characteristic times of other microphysical processes, including droplet evaporation. During this time the DSD changes substantially, especially at small cloud fraction. The 656 effective radius in the initially dry volume increases much faster than LWC. As a result, 657 effective radius in such volume rapidly reaches the values typical of cloudy air, while LWC 658 still remains substantially lower than in the cloudy volume. Despite some DSD broadening, 659 the final DSDs in the mixing volume resemble those in the initially cloud volumes. The main 660 effect of mixing is lowering the DSD values as the cloud fraction decreases. 661

662

663 **4.** Equilibrium state and mixing diagram

Mixing considered in the present study always leads to the equilibrium state. As was explained above, two equilibrium states are possible. The first one is characterized by the total evaporation of cloud droplets $q_w(x,\infty) = 0$, whereas the second one occurs if the air in the mixing volume becomes saturated, i.e. when $S(x,\infty) = 0$. At the given initial values of q_{w1} in the cloud volume and of S_2 in the initially dry volume, there always exists the cloud fraction μ_{cr} (Eq. 11) separating these two states.

670

671 **4.1.** The process of achieving the equilibrium state

Figure 7 shows the dependences of the time required to reach the equilibrium on the cloud
fraction, at different initial relative humidity values in the dry volume and two initial DSDs
(the parameters are presented in Tab.1). The characteristic time is defined here as the time

675 from the beginning of mixing to the time instance when inequality $\delta = \frac{\overline{N}(t) - \overline{N}(\infty)}{\overline{N}(0) - \overline{N}(\infty)} < 0.01$

becomes valid. The mean droplet concentration is calculated by averaging along x-axes

677
$$(\bar{N}(t) = \frac{1}{L} \int_{0}^{L} N(x,t) dx$$
). In case of a total evaporation, $\bar{N}(\infty) = 0$

678

679 Figure 7 here

680

681 Each curve in Fig. 7 consists of two branches. The left branches correspond to the total evaporation regime, while the right branches correspond to the partial evaporation at 682 equilibrium. The maximum time corresponds to the situation when the available amount of 683 684 liquid water is approximately equal to the saturation deficit. A similar result was obtained in 685 Pt1 and Pt2 for homogeneous mixing. The maximum values of the characteristic time are about 4 min for a mixing volume of 40 m in length. The right branches show that the 686 687 characteristic time decreases with increasing cloud fraction. Despite some differences in the curve slopes, the characteristic times for wide and narrow DSD are quite similar. 688

689 Figure 8 shows dependences of normalized cube of the effective radius on the cloud 690 fraction at different time instances for two values of x: x=0 (solid lines) corresponds to the initially cloudy volume, and x = L (dashed line) corresponds to the initially dry volume. The 691 figure is plotted for the narrow DSD for two values of RH_2 : 60% and 95%. Despite the fact 692 that the diffusion-evaporation equation allows simulating using any initial RH, we do not 693 consider in our examples the cases of very low RH of dry volume. It is because at very low 694 RH, say, RH=20%, the cloud fraction should exceed 0.8 to prevent total droplet evaporation 695 in the equilibrium state (at LWC=1 g/kg). We believe that turbulence above the stratocumulus 696 cloud top is very weak and entrainment is slow, so cloud fraction should be large in this case. 697 At the lateral edges of warm Cu a shell of humid air arises around cloud, so RH of the 698

entrained air should be high enough (e.g. Gerber et al., 2008).
700
701
702
703 Figure 8 here

704

The curve plotted for the time instance of 300 s corresponds to the equilibrium state (hereafter the equilibrium curve). The curves above the equilibrium curve correspond to the initially cloudy volume, and the curves below the equilibrium curve correspond to the initially dry volume. One can see how curves of both types approach the same final state. During the

709 mixing the curves move over the $\left(\frac{r_e}{r_{e0}}\right)^3 - \mu$ plane toward the equilibrium curve. As a result,

the curves plotted in Fig.8, corresponding to different time instances of the mixing, togethercover the entire area of the panels.

712 During this movement the distance from the curves to the horizontal line $\left(\frac{r_e}{r_{e0}}\right)^3 = 1$ changes,

and the curves slopes increase. In our case of L = 40 m, the mixing remains inhomogeneous the during entire mixing process, so the change in the distance from the curves to the horizontal line $\left(\frac{r_e}{r_{e0}}\right)^3 = 1$ characterizes the temporal changes over the mixing process, but not

716 a change in mixing type.

717 It is noteworthy in this relation that scattering diagrams plotted using *in-situ* observations 718 reflect mixing between different multiple volumes at different stages of the mixing process. 719 Accordingly, points in the scattering diagrams can be far from the equilibrium location. Fig. 8 720 indicates, therefore, that scattering diagrams show snapshots of transient mixing process when 721 the distance from points in the diagrams to line $\left(\frac{r_e}{r_{e0}}\right)^3 = 1$ characterize the stage of the

mixing process, but not the mixing type.

time instances at wide DSD also indicate approaching to the equilibrium curve, while all the

The dependences of normalized cube of the effective radius on the cloud fraction at different

r25 curves correspond to $\left(\frac{r_e}{r_{e0}}\right)^3 > 1$ (not shown). In most studies normalized cube of mean r26 volume radius is used for plotting the mixing diagrams. Note, however, that since effective r27 radius in a wide range of conditions is only by 10% larger than the mean volume radius, the r28 utilization of effective radius does not affect the results. Moreover, satellites measure r29 specifically effective radius. Accordingly, some authors (e.g. Freud et al., 2011) use effective r20 radius for plotting the mixing diagrams.

731

723

732 **4.2.** Mixing diagrams

Using the diffusion-evaporation equations (15-17) we calculated the equilibrium DSD for different initial relative humidity values and different cloud fractions. Each calculation was performed for both narrow and wide initial DSD (parameters shown in Tab.1). These equilibrium DSD were used to calculate mixing diagrams showing dependences of normalized cube of the effective radius on the cloud fraction.

The corresponding mixing diagrams for homogeneous mixing case were also calculated for comparison. To this effect, the supersaturation and DSD in both the cloud and the dry volumes were aligned, taking into account the cloud fraction value μ . The alignment led to the following initial values of supersaturation and DSD within the mixing volume:

742

743
$$S_0 = (1 - \mu)S_2; \quad g_0(\sigma) = \mu g_1(\sigma)$$
 (21)

⁷⁴⁵ Upon the alignment, time evolution values of DSD under homogeneous evaporation in an ⁷⁴⁶ adiabatic immovable parcel were calculated until the equilibrium state was reached. These ⁷⁴⁷ equilibrium DSD were used to calculate mixing diagrams for homogeneous mixing. To do ⁷⁴⁸ this, we used the parcel model proposed by Korolev (1995) that describes evaporation by ⁷⁴⁹ means of equations with temperature-dependent parameters. **Figure 9** shows the mixing ⁷⁵⁰ diagrams plotted for initial narrow and wide DSD cases.

- 751
- 752 Figure 9 here
- 753
- 754

While all the curves in the mixing diagram for narrow DSD are below the straight line 755 $\left(\frac{r_e}{r}\right)^3 = 1$, the curves for wide DSD are above this line. The explanation of this effect is given 756 in Section 3 (Fig. 6). The curves plotted for homogeneous and inhomogeneous mixing 757 demonstrate an important feature. Namely, at given values of RH and q_{w1} in the initially dry 758 volume, the values μ_{cr} of the cloud fraction at which all the droplets evaporate are 759 approximately the same for any type of mixing. This condition is the consequence of the mass 760 conservation law determined by Eq. (11) and does not depend of the initial DSD shape. In 761 standard mixing diagrams (e.g. Lehmann et al., 2009; Gerber et al., 2008; Freud et al., 2011), 762 the horizontal straight line $\left(\frac{r_e}{r_{e0}}\right)^3 = 1$ is typically plotted for the entire range of the cloud 763 fraction [0...1], while the curves corresponding to homogeneous mixing are plotted for 764 different RH within the range $[\mu_{cr}(RH_2)...1]$. As a result, the high difference between 765 extremely inhomogeneous and homogeneous mixing types is clearly seen at low RH and at 766 small cloud fractions. The condition that μ_{cr} is the same for different mixing types indicates 767 that the mixing diagrams may look nearly similar for $\mu > \mu_{cr}$. It means that the range of the 768

cloud fractions required for comparison of diagrams aimed at determination of a mixing type shortens as RH_2 values in the surrounding air decrease.

The comparison of the left and the right panels in Fig. 9 shows that the differences 771 772 between the diagrams for homogeneous and inhomogeneous mixing types are more 773 pronounced for initially narrow DSD. The maximum difference should take place for monodisperse DSD considered in in Pt1, Pt2 and Pt3. Within the range of $\mu > \mu_{cr}$, the 774 775 distance between the curves corresponding to different mixing regimes is small even for narrow DSD and low RH_2 . The lower difference is related to the fact that at high RH_2 the 776 777 curves in the mixing diagrams are close to the horizontal straight line in both regimes, while at low RH_2 , μ_{cr} is small and both curves should drop to zero in the vicinity of $\mu = \mu_{cr}$. 778

As regards the wide DSD case, the difference between the curves corresponding todifferent mixing type is negligible (Fig. 9, right)

781

782 **4.3. Effect of the relative humidity**

783 In measurements carried out at cloud boundaries and in cloud simulations, the cloud fraction is not known, therefore it is widely accepted to use normalized droplet concentration 784 785 instead of the cloud fraction (Burnet and Brenguier, 2007; Gerber et al., 2008: Lehmann et al., 2009). Droplet concentration is normalized by the maximum value along the airplane traverse. 786 787 The difference between the cloud fraction and normalized droplet concentration is obvious: 788 the cloud fraction is a parameter given as the initial condition. At the same time, normalized droplet concentration changes with time and space due to complete evaporation of some 789 droplet fraction. Figure 10 shows dependencies of normalized droplet concentration on the 790 791 cloud fraction at the equilibrium final state of mixing. One can see a substantial deviation from 1:1 linear dependence, especially at low RH. As we know, droplet concentration 792 793 decreases in the course of both homogeneous and inhomogeneous mixing if the initial DSD are polydisperse. The fraction of totally evaporating droplets increases with decreasing RH_2 . 794

As expected, droplet concentration in homogeneous mixing is higher than that in
inhomogeneous mixing. The difference between droplet concentrations at wide DSD is lower
than at narrow DSD.

798

Fig. 10 here

800

Figure 11 shows the dependencies $\left(\frac{r_e}{r_{e0}}\right)^3$ on normalized droplet concentration for narrow 801 and wide DSD in inhomogeneous mixing. The normalization by droplet concentration in the 802 initially cloud volume at t = 0 was used. Taking into account the dependences of normalized 803 droplet concentration on the cloud fraction μ (Fig. 10), one can get the curves shown in Fig. 804 11 which actually coincide at different RH_2 . The lack of the sensitivity to RH_2 can be 805 attributed to the fact that a decrease in RH leads to a decrease in normalized droplet 806 807 concentration, so the curves corresponding to low RH in Fig. 9 shift to the left when the normalized droplet concentration is used instead of μ . The shape of the dependences in Fig 808 11 (right) is explained by an increase in the effective radius with decreasing droplet 809 concentration. 810

811

812 **Fig 11 here**

813

814

Thus, the mixing diagrams plotted in the plane $\left(\frac{r_e}{r_{e0}}\right)^3$ vs normalized droplet concentration do not depend on the relative humidity of the surrounding dry air. This result

concentration do not depend on the relative humidity of the surrounding dry air. This result
indicates an additional difficulty in distinguishing between mixing types based on scattering
diagrams plotted using *in-situ* data in these axes. The concentration of observed points in

818 these scattering diagrams close to the line $\left(\frac{r_e}{r_{e0}}\right)^3 = 1$ is often interpreted as an indication of

homogeneous mixing, but at high RH in the surrounding air (Gerber et al., 2008; Lehmann et
al., 2009). High values of RH in the penetrating air volumes are usually explained by
formation of a layer of most air around the cloud boundary (Gerber et al., 2008, Knight and
Miller, 1998).

The reference values of droplet concentration and the effective radius used for normalization in the present study are taken as the initial values in the cloud volume before it mixes with the neighbouring dry volume. In real *in-situ* measurements the reference values of these quantities are typically chosen in a less diluted cloud volume along the airplane traverse. This reference volume may be quite remote from the particular mixing volume. It can lead to a shift of the mixing diagram with respect to the $\left(\frac{r_e}{r_{e0}}\right)^3 = 1$ line, as well as to a large variation

829 in mixing diagram shapes, unrelated, however, to the mixing type (e.g., Lehmann et al.,830 2009).

831

832 5. Discussion and conclusion

This study extends the analysis of mixing performed in Pt3 where the 833 diffusionevaporation equation served as the basis, the initial DSD were assumed monodisperse and 834 the cloud fraction was chosen as $\mu = 1/2$. In the present study, the analysis focuses on the 835 temporal and spatial evolution of initially polidisperse DSD and investigates mixing diagrams 836 obtained for narrow and wide initial DSD within a wide range of the cloud fraction values (837 0.1 - 0.95). It is shown that results of mixing and the structure of mixing diagrams depend on 838 the initial DSD shape. This finding indicates that mixing is a multi-parametrical problem that 839 cannot be determined by a single parameter (e.g. the Damkölher number as often assumed) or 840 even by two parameters (the Damkölher number and the potential evaporation parameters as 841

assumed in Pt3). The temporal changes of DSD and their moments during mixing are 842 calculated. Although DSD broaden, they tend to remain similar to the original DSD. The main 843 changes come from the cloud air dilution by the dry air, which leads to a decrease in droplet 844 concentration for all droplet sizes. The changes of DSD and its shape are minimum in the 845 initially cloud volumes, especially at significant cloud fractions. The droplet radii 846 corresponding to the DSD peak do not change anyhow significantly. In the initially dry 847 volumes, mixing leads to a rapid increase in RH. Consequently, large droplets penetrating 848 these volumes do not change their sizes significantly. As a result, the effective radius in these 849 volumes rapidly increases and reaches the values typical of cloud volumes, while LWC 850 851 remains lower than in the cloud volume for most of the mixing time. At narrow DSD, the effective radius remains smaller than that in the initially cloud volume. At wide DSD, the 852 effective radius may become larger than that in the initial DSD. This increase in the effective 853 854 radius is attributed to the fact that evaporation of smaller droplets leads to the increase in the fraction of larger droplets in the DSD. Note that the DSD broadening caused by mixing is the 855 consequence of the utilization of polydisperse DSDs. In classical theory that assumes 856 monodisperse DSDs no broadening takes place. This problem is analyzed in detail in Pt 3. 857 Note that in real clouds DSD there are many mechanism leading to DSDs broadening (e.g. 858 859 Pinsky and Khain 2002).

Be Dependences of normalized cube of the effective radius on the cloud fraction $(r_e / r_{e0})^3$ as a function of μ at different time instances form the set of curves filling the entire $(r_e / r_{e0})^3 - \mu$ plane. Therefore, both the slope and the distance of these curves in respect to the horizontal line $(r_e / r_{e0})^3 = 1$ change with time. It means that this distance characterizes the temporal changes in the course of mixing, but not the mixing type (which remains inhomogeneous during the entire mixing time). The mixing process is comparatively long (several minutes), so the final equilibrium stage is hardly achievable in real clouds. It is highly significant that the critical values of the cloud fraction μ_{cr} corresponding to total droplet evaporation are the same for any mixing type. It means that the curves in a mixing diagram corresponding to homogeneous and inhomogeneous mixing types should be compared only within the range of $\mu > \mu_{cr}$. The range width of $\mu > \mu_{cr}$ decreases with decreasing relative humidity in the initially dry volume. Taking into account significant scattering of observed points, this condition greatly hampers the problem of how to distinguish between mixing types,

Another important result of the study is that mixing diagrams for homogeneous and inhomogeneous mixing plotted for polydisperse DSD do not differ much. The largest difference takes place for initially narrow DSD (the maximum difference should take place for initial monodisperse DSD), but even in this case the difference is not large enough to reliably distinguish mixing type due to the significant scatter of observed data. At wide DSD, this difference becomes negligibly small.

The cloud fraction μ is a predefined parameter and is not determined from observations. 880 881 Consequently, in the analysis of *in-situ* measurements the normalized droplet concentration is typically used instead of the cloud fraction. However, there is a significant difference between 882 the cloud fraction prescribed a priori and the normalized droplet concentration that changes 883 due to total evaporation of some fraction of droplets. We have shown that utilization of 884 normalized droplet concentration in mixing diagrams is not equivalent to utilization of the 885 cloud fraction. The important conclusion is that when mixing diagrams are plotted using the 886 normalized concentration, the dependence on the RH disappears. This conclusion is valid even 887 when the RH in the initially dry volume is as low as 60%. This conclusion clearly contradicts 888 the wide-spread assumption that mixing types can be easily distinguished in mixing diagrams 889 in case of low relative humidity of the surrounding air. 890

To sum up, our general conclusion is that the simplifications underlying the classical concept of mixing are too crude, making it impossible to use mixing diagrams for same time, mixing diagrams may contain useful information on DSD width.

895

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904 Appendix. List of symbols

Symbol	Description	Units
<i>A</i> ₂	$\frac{1}{q_v} + \frac{L_w^2}{c_p R_v T^2}$, coefficient	-
a_n	Fourier series coefficients	-
С	Richardson's law constant	-
C _p	specific heat capacity of moist air at constant pressure	J kg ⁻¹ K ⁻¹
D	coefficient of water vapor diffusion in air	$m^2 s^{-1}$
Da	Damkölher number	-
е	water vapor pressure	N m ⁻²
e_w	saturation vapor pressure above flat surface of water	N m ⁻²
F	$\left(\frac{\rho_w L_w^2}{k_a R_v T^2} + \frac{\rho_w R_v T}{e_w (T) D}\right), \text{ coefficient}$	m ⁻² s
f(r)	droplet size distribution	m ⁻⁴
g(r)	droplet size distribution	m ⁻⁵

$g_0(\sigma)$	initial distribution of square radius in homogeneous mixing	m ⁻⁵
$g_1(\sigma)$	initial distribution of square radius	m ⁻⁵
<i>k</i> _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
Ν	droplet concentration	m ⁻³
N_0	Parameter of Gamma distribution	m ⁻³
\overline{N}	mean droplet concentration	m ⁻³
N_1	initial droplet concentration in cloud volume	m ⁻³
р	pressure of moist air	N m ⁻²
$q_{_{v}}$	water vapor mixing ratio (mass of water vapor per 1 kg of dry air)	-
$q_{\scriptscriptstyle w}$	liquid water mixing ratio (mass of liquid water per 1 kg of dry air)	-
$q_{\scriptscriptstyle w1}$	liquid water mixing ratio in cloud volume	-
R	$\frac{S_2}{A_2 q_{w1}}$, non-dimensional parameter	-
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
r	droplet radius	m
r_1	initial droplet radius	m
r _e	effective radius	m
r_{e0}	initial effective radius	m
S	$e/e_w - 1$, supersaturation over water	-
S_2	initial supersaturation in the dry volume	-

S_{0}	initial supersaturation in homogeneous mixing	-
Т	temperature	K
t	time	S
x	distance	m
α	parameter of Gamma distribution	-
β	parameter of Gamma distribution	m ⁻¹
Δt	time step	S
μ	cloud fraction	-
μ_{cr}	critical cloud fraction	-
ε	turbulent dissipation rate	m^2s^{-3}
$\Gamma(x,t)$	conservative function	-
$ ho_a$	air density	kg m ⁻³
$ ho_{\scriptscriptstyle W}$	liquid water density	kg m ⁻³
σ	square of droplet radius	m ²
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Tab.1 Parameters of the initial Gamma distributions

	DSD	N_0 , cm ³	α	eta , $\mu \mathrm{m}$	Modal radius, μm	LWC, g/m ³
	Narrow	264.2	101.0	0.1	10.0	1.178
	Wide	71.0	4.3	3.1	10.0	1.178
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1088 Fig. 2. Spatial-temporal variations of conservative function $100 \times \Gamma(x,t)$ for different cloud 1089 fractions μ and initial $RH_2 = 80 \%$.



Fig. 3. Time evolution of DSD in the centers of the initially cloudy volume (left) and of the initially dry air volume (right) at initially narrow DSD. The initial mixing parameters are $RH_2 = 80 \%$, $T = 10^{\circ}C$, p = 828.8 mb and L = 40 m.







1145 Fig. 5. Spatial dependences of droplet concentration, LWC and the effective radius within the 1146 mixing volume at different time instances at narrow initial DSD. The initial mixing 1147 parameters are $RH_2 = 80 \%$, $T = 10^{\circ}C$, p = 828.8 mb and L = 40 m.





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Fig. 8. Dependences of normalized cube of the effective radius on the cloud fraction at different time instances for x=0 (solid lines) corresponding to the initially cloud volume, and x = L (dash line) corresponding to the initially dry volume. The time instances in seconds are marked by numbers. The figure is plotted for the narrow initial DSD for two values of RH_2 : 60% (left panel) and 95% (right panel). Parameters of DSD are given in Tab. 1. The initial mixing parameters are $T = 10^{\circ}C$, p = 828.8 mb and L = 40 m. Calculations performed within the range of $0.1 < \mu < 0.95$.

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Fig. 9. Mixing diagrams. Normalized cube of the effective radius vs. the cloud fraction for initial narrow DSD (left) and initial wide DSD (right). The dependencies correspond to the equilibrium state Parameters of initial DSD are presented in Tab. 1. Solid and dashed lines show the mixing diagrams for inhomogeneous and homogeneous mixing, respectively. The initial mixing parameters are $T = 10^{\circ}C$, p = 828.8 mb and L = 40 m.

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Fig. 11. Dependencies of normalized cube of the effective radius on normalized droplet concentration for different initial relative humidity values. Left panel: narrow initial DSD. Right panel: wide initial DSD. The initial mixing parameters are $T = 10^{\circ}C$, p = 828.8 mb and L = 40 m.