Referee report.

We are very appreciate to both referees for the referee reports, allowed us to improve the manuscript. Please, find below the detailed answers on both reports.

Referee 1

This article studies the effect of aerosol dynamics on atmospheric small-scale turbulence using direct numerical simulations.
As I already pointed out in my original assessment of the article, my main concern with this article are the extreme initial conditions that were chosen for the simulations: While I understand the concept of fluctuations and the concurrent possibility of achieving extreme values, it is very hard for me to assess how relevant it is to study such an extreme case outside of that context. To elaborate on what I mean, let's take the article by Kulmala et al. that has also been cited by the authors: Kulmala et al. Treat the saturation ratio (let's call it S_0 here, because S is used for the supersaturation in the present article) as a stochastical variable with a Gaussian distribution around an average value which varies from 0.995 to 1.0 with a standard deviation of up to 0.05. They then conduct a series of simulations where they allow the saturation ratio to vary randomly according to the assumed distribution and find that particles can activate also in under-saturated conditions due to the temporal fluctuations in the saturation ratio. In the present paper, the authors pick one very extreme case out of this distribution, which corresponds to a saturation ratio of 1.1 or a supersaturation of 10 % (I can only guess that they still assume the average S_0 in the cloud to be equal to one). Just to put this into context, common supersaturation values at the base of a cloud are of the order of 0.1 to 0.5 %; according to a quick test conducted with a cloud parcel model that does not consider fluctuations, it reqires a particle number concentration of 1 cm^{-3} and an updraft velocity of 10 m/s to achieve a supersaturation of 10 %. Furthermore the authors chose a very high temperature difference between the simulation domain and its surroundings, which is not motivated in the text at all. According to these extreme initial conditions, the authors then also find that aerosols have a strong influence on turbulence, but I wonder how justifiable such a conclusion is without also considering more moderate supersaturations which are, after all, much more likely to occur. Furthermore, I am a little bit skeptic how reliable the results presented here are, as the simulations include the use of random numbers (this especially concerns the generation of the initial turbulence) and thus a single simulation may not be very representable of an average behaviour.

To conclude, I cannot recommend this article for publication in the current form. At the very least the paper requires one more set of reference simulations with a more conventional supersaturation of, say, 0.3 %, and a proper discussion on the issues I layed out above.

According to the referee's comment we have made three additional runs

- the difference between the temperature in the simulation domain and its surroundings of 2.5 K and aerosol particles are included

- the difference between the temperature in the simulation domain and its surroundings of 2.5 K and aerosol particles are not included

- humidity is 10 % smaller than it was in the previous simulations and therefore, supersaturation averaged over domain is 0.6

The last section about the effect of aerosol on the turbulent motion is rewritten based on new input parameters. And a new subsection about effect of initial supersaturation on activation of aerosol

particles is included

Concrete remarks

1. The English is not very good and needs to be reviewed. Some of the sentences are very hard to understand.

We double checked the English and rewritten unclear parts.

2. lines 12–14: Latent heat release is a time dependent process, but finally, close to equilibrium, the total cooling depends (nearly) only on the initial amount of supersaturation. It is therefore unnecessary to state that "even small amounts of aerosols increase the air temperature", and it is quite misleading to give the (very high) change in temperature of 1 K without also giving the value for the supersaturation used.

Reformulated:

We find that the even small amount of aerosol particles (55.5 cm^{-3}) strongly affects the air temperature due to release of latent heat.

3. lines 115–120: Why is the chemical composition of the air important to this study? Wouldn't it be enough to state the total water content?

The model used in this study was prepared for problems with more complicated chemistry. We are planning to develop it in future.

4. lines 126–129: Is this the total difference in temperature/water vapor content between the (upper and lower?) edges of the domain (e.g. 0.001 K/10 cm)? According to the units it cannot be a gradient.

Reformulated:

Based on data of CARRIBA observations typical for the upper parts of clouds / cloud edges at a height of 2000 m, we set the initial conditions for air temperature (T0 = 285.4 K) and water vapor mixing ratio (q0= 0.0124). The small vertical gradients of temperature and water content are also based on the CARRIBA measurements: the total difference between values of air temperature and water vapor mixing ratio at the upper and lower edges of the domain are Delta T = 0.001 K and Delta q= 4 10^{-5}, correspondingly.

5. lines 133–135: How exactly was this range of supersaturation derived?

The take measurements for temperature and absolute humidity (AH).
S=(AH*rho*pw/ps-1)*100%

AH=Yw/rho
Yw is water vapour mass fraction
rho is air density
pw=rho RT/18 is water vapour pressure

psf is saturation water vapour pressure

rho=9.6 10^{-4} g/cm3
Yw= 0.0123759
T=285.4 K

6. line 264: I have never heard the word “unequilibrium” before – I believe the correct term is “non-equilibrium”.

We agree that the phrase “non-equilibrium” is more common and we change accordingly

7. Figure 1: The velocity fields in panels (a) and (c) look curiously similar, while they are totally different in panel (b). Did you use exactly the same initial velocity fields in all three cases (or probably a different one for N_{tot} = 555 cm^{-3})? How much does varying the particle concentration actually affect turbulence?

Velocity field is taken the same in all free cases. Panels (a) and (c) look similar because the positions of warm/cold layer are similar there (comparing with panel (b)). But it was double checked that the difference between (a) and (c) exists.

8. lines 306–317: How does the initial supersaturation vary? Are these the values that you mention in lines 133–135? If so, it is not obvious how only the temperature is responsible for this variation in supersaturation, as you also set a gradient in water vapour mixing ratio. I think a figure that clearly lays out the initial conditions of the simulations would be very helpful in the overall understandability of this paper.

In the first version of the manuscript such figures were included but the other referee asked to remove them because in his opinion they were not informative.

9. lines 323–326: Can you somehow quantify how long it would take the system to equilibrate, “more than 3 s” is pretty vague?

We added a new figure to show time dependence of supersaturation averaged over domain for Ntot=55.5, 555 and 5550.

In this Fig. we present the supersaturation averaged over domain for cases 1, 2, 3 and 4 (see Table 1) and analyze the relaxation time of supersaturation for different values of initial supersaturation and total number of particles. Analyzing numerical results we get the relaxation time of about 0.77 s (case 2), 0.17 s (case 3), and 0.6 s (case 4). In case 1 the relaxation time is larger than time of simulations. We fit the corresponding curve by the function exp(-t/tau) and estimate the relaxation time of 4 s.

10. lines 341–347: As the condensational growth of particles is limited by the total surface area, it is not very surprising that the water vapor mixing ratio decreases faster if more particles are present. Also, as for this reason the supersaturation is always larger for smaller number concentrations, it should be

expected that these particles grow larger. I don't know if this really "confirms the Twomey effect" or if it rather confirms that the model works as it should.

Removed the sentence about Twomey effect

11. line 359: Replace "number" with "fraction".

replaced

12. Figure 5: Why does the fraction of activated particles go down after about half a second for N_{tot} = 55.5 cm^{-3}?

This is most probably the effect of nonlinearity of activation process and periodic BC.
There are no such fluctuations in other two cases because in these two cases the system equilibrates very fast.

13. lines 380–384: I don't understand the relevance of this statement. As you have the same total water content in all three cases to begin with, the difference in LWC very strongly depends on the time at which you compare the simulations (apart from some small corrections due to temperature and droplet size), because in some simulations the droplets still grow while in others they don't. How would this difference change if you waited 6 instead of 3 seconds?

Added to the text that this result depends on time

We find that at t= 3 s the number of activated particles is proportional to the total number, whereas the change of Ntot by a factor of hundred increases LWC by approximately 40 %(Table3).

14. lines 385–399 and Figure 6: How exactly does the buoyancy force affect the turbulence in the simulation domain at scales smaller than 10 cm? This is not discussed at all in the manuscript. How valid is the assumption of a temperature difference of 7.6 K between the simulation domain and its surroundings, especially when the cloud parcel is colder than the rest? From Figure 6 I roughly estimate a deceleration of the air parcel of 25 cm s^{-2} (solid black line), so the air parcel drops more than 1 m (more than ten times the domain size) during the 3 s simulation, still the environment temperature never changes. How does this conform with the original statement that the temperature in the cloud is subject to strong fluctuations? On a side note, if the temperature changes by 1 K, it is not surprising that the buoyancy force changes quite a bit and I wonder if Figure 6 is necessary at all – why not just give a value for B in the different cases? Furthermore, what does the comparison of the two simulations tell us. No aerosols means no cloud – should you rather compare between different aerosol loadings to somehow mimic what could happen at the edge of the cloud?

As it was mention at the beginning of this respond we have prepared results of simulations in a case of smaller difference between the temperature inside the domain and its surroundings (delta T= 2.5 K). Moreover, now the temperature inside the domain is larger than outside the domain and we get updraft instead of downdraft. We agree that new parameters are more realistic in atmosphere.
However, our general conclusion concerning the effect of aerosol on turbulence does not change. We showed in the last section that the aerosol affects the turbulence through the buoyancy.

However, since now temperature difference changes sign the vertical air motion is accelerated (rather than decelerated as it was before) if aerosol particles are present.

15. lines 400–410: To my eye the TKE in Figure 7 only differs significantly between aerosols and no aerosol during a short period of time around 2 s. Accordingly, Figure 8 shows a strong jump from roughly zero up to 80 % at that point in time and starts dropping off again thereafter. Still, from Figure 8 the authors conclude that aerosols affect turbulence strongly. How sure are you that these results are not sensitive to the specific set of random numbers used for the simulation (i.e. what would happen if you used a different random seed in the setup)?

We agree that to make the general conclusions about the effect of the turbulence one set of parameters are not sufficient, and since the turbulence is very complicated process some statistics is needed. But the DNS are very computationally demanded and even one set of parameters needs huge amount of time and resources. In this study we illustrate how aerosol important or not important for correct description of turbulent motion, taking conditions typical for atmosphere.

16. lines 406–410: This sentence is very hard to understand. Do you mean to say that the temperature difference between the domain and its surroundings decreases because the temperature inside the domain increases?

Yes.

But now we take warmer domain and cooler its surroundings and instead of decrease/deceleration we get increase/acceleration effect.

This part is reformulated:

The air temperature increases because of release of latent heat caused by condensation onto droplets, and therefore, the difference between temperatures inside and outside the domain is enlarged. It results in increasing of buoyant force and accelerating of air motion in vertical direction.

Referee 2.

General:
The study on aerosol particle dynamic effects is a spectacular idea and performance on small scale variation effects on cloud properties such as activated particles and temperature effects usually either ignored or simply parameterized. The approach and the implications for example for larger scale aerosol particle – cloud effect calculations matches nicely in the scope of Atmospheric Chemistry and Physics and the results are quite interesting. However before accepting the present study I would recommend several technical improvements and clarifications in order to support readers not essentially familiar with all the details to follow the arguments and the implications for larger scale simulations. Those include first of all the English. Please have a native English speakers check on the sentences!

• The total number of particles was varied between two orders of magnitude, which was extracted from reasonable values measured. This is

appropriate and reasonable. However, what about the impact of different size ranges e.g. mode concentrations on the results? Do the results change notably for particles in the accumulation and in the coarse mode due to critical sizes for activation for the salt particles assumed? Would results differ for changing certain size bin concentrations (i.e. modes) instead of the whole number? Are these salt particles already "activated" or assumed "dry" for the simulations conducted? I guess once any of these particles has faced substantial humidity it will grow much easier than if it has to dissolve first.

These are very interesting questions and we would definitely like to perform more simulations to answer, but one model simulation for 3s takes about 2 weeks at a cluster machine when using 512 cores. So the computational costs are very high for these runs and for this reason it was not possible to perform runs with e.g. different size bin distributions.

Evolution of the particle distribution function was accurately tested in previous study. (Babkovskaia et al. 2015). The minimal size bin and the time step are chosen such that for one time step the particles move to the neighbor bin. We checked that the increasing/decreasing of the bin size and corresponded decreasing/increasing of the time step do not make any effect on the final result. Also, we assume that initially the particles are almost dry, i.e. there is a solid core with very thin water envelope to start the activation.

• Abstract, p.1: "The system comes to an equilibrium faster and the relative number of activated particles appears to be smaller for larger N_{tot}." seems to be formulated very simple. I doubt that for a large part of atmospheric processes equilibrium conditions are hardly reached. What is the criteria for achieving an equilibrium condition in this case and for which simulation conditions the equilibrium approach becomes invalid?

Indeed, the phase relaxation time is $tau_{phase} \sim (Ntot \langle r \rangle)^{-1}$, where $\langle r \rangle$ is the mean droplet radius. The steady-state supersaturation can be written as $Sqs \sim a1 \ tau_{phase}$, where a parameter $a\_1\ m^{-1}$ is a parameter including thermodynamic parameters and being almost constant. Thus, it becomes clear that for larger Ntot the system comes faster to an equilibrium.

• Description of the model, p. 2: The order of figures seems somewhat arbitrary, as Fig. 3 appears earlier than Fig. 1.

Fig 3 is moved to the beginning of section 2

• p. 2, l. 106ff: The particle size distribution displays a sharp maximum close to a diameter of around 5 micron. Please refer to the origin of observations (reference, location etc.) mentioned in the text.

In the caption of Fig 3 it was mentioned that dotted curve corresponds to the observed distribution of aerosol, and solid curves are distributions of droplets. There are no measurements for 5 micro-meter particles.

• p. 2, l. 127: It's being referred to a temperature gradient of 0.001 K. Two questions on that: (i) which gradient, i.e. temperature change over which distance, horizontal, vertical etc.? Only a temperature unit is provided. (ii)

This temperature change is pretty tiny although important. What is the reliability range of this because of numerical diffusion and linearization of equations for simulation? Please provide a temperature gradient and either a short statement of simulation uncertainty or a value.

This part was rewritten:

This model represents the 3D fluid flow on the microscale inside a volume of 10 cm x 10 cm x 10 cm, just inside the cloud in the mid-troposphere. Based on data of CARRIBA observations typical for the upper parts of clouds / cloud edges at a height of 2000 m, we set the initial conditions for air temperature (T_0 = 285.4 K) and water vapor mixing ratio (q_0= 0.0124). The small vertical gradients of temperature and water content are also based on the CARRIBA measurements: the total difference between values of air temperature and water vapor mixing ratio at the upper and lower edges of the domain are Delta T = 0.001 K and Delta q= 4 \times 10^{-5}, correspondingly.

• p. 3, Fig. 2: I do understand the intention to maximize differences in the color scale to make aspects visible. However, since in here three situations are compared with, please use the same scale for all the three upper and all the three lower plots. This would allow a better comparison and an even improved identification of the changes.

The differences between maximal and minimal values in the corresponding cases are much smaller than the difference between plots. Keeping the same max and min for all three cases we could not resolve the distribution of temperature (supersaturation) in one case. We have left the plots in previous form.

• p. 3, l. 163ff: Please reformulate: “: : : and the usual equilibrium supersaturation would be restored.”. I doubt an equilibrium supersaturation, as water tends to equilibrate at saturation. If you mean different, please reformulate to make it clearer.

Reformulated:

On the other hand, the supersaturation excess would be eliminated by condensation onto droplets and quasi-steady state supersaturation would be restored

• p. 3, l. 172ff: Please check: “If the phase relaxation : : : would be applicable.” There seem to be too many words. Is the word “than” dispensible?

Fixed:
If the phase relaxation time is smaller than the turbulent mixing time then the actual supersaturation will tend to the quasi steady-state solution.

• p. 4, l. 229: You state that the number of particles stays constant. This contradicts the explanation of an aerosol particle dynamic study. Are changes if calculated in the corresponding simulation time negligible? Otherwise this may matter as e.g. larger cloud droplets grow on the expense

of smaller droplets and they modify the size spectrum and number density.

Here we mean the total (i.e. integrated over all sizes) number of particles in the domain. The periodic boundary conditions means that the particles can not come in/out from the domain.

• p. 4, l. 266 and p. 2, Table 2: The change in temperature between equilibrium and unequilibrium case seems fairly huge! 8K would cause a strong vertical uplift, a strong local mixing (dilution), which would require a remarkable mass of condensed water vapor (several grams per m3). Did I get something wrong?

We have prepared results of simulations in a case of smaller difference between the temperature inside the domain and its surroundings (delta T= 2.5 K). Moreover, now the temperature inside the domain is larger than outside the domain and we get updraft instead of downdraft. We agree that new parameters are more realistic in atmosphere. However, our general conclusion concerning the effect of aerosol on turbulence does not change. We showed that the aerosol affects the turbulence through the buoyancy. However, since now temperature difference changes sign the vertical air motion is accelerated (rather than decelerated as it was before) if aerosol particles are present.

• p. 4, l. 278f: The temperature is averaged in y-direction. If you have notable differences in x- and z-direction, how does this assumption affect the results? To a negligible extend?

The notable gradients of corresponding variables exist only in vertical direction (z). In x- and y-directions the changes are only because of turbulent fluctuations (not so strong). Therefore, averaging in y- direction does not make any crucial effect on the results.

• p. 5, Table 3: I don't understand the listed maximal and minimal values of supersaturation S as they are negative. This would imply a subsaturation as S = 1-p/psat0 with p and psat0 the vapor pressures of water at present and at saturation level. Second, very interesting is the change between cases 1 and 2. There seems to be a tipping point at a certain total particle number concentration. Could you provide a comment on that as the changes by a factor of ten is substantial?

We introduce supersaturation as S=p/psat-1. It decreases with time. Therefore, the difference between values at t=3 s and t=0 s is negative. The caption is rewritten:
Initial value of supersaturation averaged over the domain \overline{S_{init}}, total number of particles (Ntot), number of activated particles (Nact) at t=3 s, liquid water content (LWC) at t=3 s, change in temperature between start and end of simulation (Delta T), change in percentage maximal supersaturation between start and end of simulation (Delta Smax), change in percentage minimal supersaturation between start and end of simulation (Delta Smin), relaxation time of supersaturation tr at t=3 s for considered cases 1, 2, 3, 4 (see Table1). In cases 2,3,4 tr is the numerically predicted value (time for 63 % change form start). In case 1 the phase relaxation time is obtained from Eq.6. No subsaturation is predicted anywhere in the domain in all cases.

Concerning the tipping point mentioned we agree with the referee that this would be an interesting point to study further and will also consider it in our next research. However, because of the high

computational costs of the simulations we decided to leave this open for the future. Speculative we would argue that at a certain number concentration the activation for a given supersaturation is not anymore limited by the amount of particles, which would mean that at this time the diffusion limitation of water molecules reaching the particles is negligible. However, to prove this statement more runs with different number concentrations and a set of different supersaturation would be required.

• p. 6, l.298f: You state that the simulated results occur because of the effect of total number concentration. Why? I guess a certain limit of aerosol particles – here all assumed to be identical in chemical composition and water solubility – exists, below which the time of diffusion of water vapor to the next aerosol particle is too long to achieve the same amount of condensation. Because of the particles size (predominantly beyond 1 micron in diameter) hardly any curvature effects on saturation vapor pressure can be expected. If so, could you name the cutting point for the conditions simulated in here?

No at this time we are not able to name the cutting point as the referee mentioned. As already explained above our hypothesis is that there is a diffusion limitation between the water molecules and the particles for certain amount of aerosol number concentrations, however, the complete study of this interesting phenomena was out of the frame of this manuscript but will be considered in our future research.

• p. 6, l. 330: The point mentioned above feeds back to the statement dealing with the activation radius assumed. Why exactly 1.75 micron? This should depend on supersaturation. “: : :the results of this study were not sensitive on the choice of “ the activation radius. My guess (!) is that this is valid for the cases 2 and 3 but not for case 1. Do you agree or disagree?

For three cases with the same initial supersaturation the results do not depend on critical radius if it is smaller than 1.75 micron. We have prepared additional run for smaller supersaturation and discussed effect of supersaturation in additional section. We find that for smaller supersaturation the system comes to an equilibrium for the same time as in a case of large supersaturation but the final size of particles are appeared to be smaller. We agree that to generalize our results for different initial parameters more simulations are needed.

• Fig. 6: The calculated vertical velocities of 0.6 to 0.7 m/s at maximum are remarkable. It is indicated that this intensifies over time although a steady-state or “equilibrium” is to be achieved after a second or somewhat more.

We have prepared results of simulations in a case of smaller difference between the temperature inside the domain and its surroundings (delta T= 2.5 K). Moreover, now the temperature inside the domain is larger than outside the domain and we get updraft instead of downdraft. We agree that new parameters are more realistic in atmosphere.
However, our general conclusion concerning the effect of aerosol on turbulence does not change. We showed in the last section that the aerosol affects the turbulence through the buoyancy. However, since now temperature difference changes sign the vertical air motion is accelerated (rather than decelerated as it was before) if aerosol particles are present.

• p. 6, Fig., 7: “The dependence of the average...” turbulent “kinetic energy: : :”. Please insert.

inserted

• p.7, 353f: Aerosol dynamics are neglected. This sounds different in the abstract as it is stated that in order “to study effects of aerosol dynamics on the turbulence we vary: : :”. Please name explicitly in the methods section not to use aerosol dynamics and state that this is valid because of the short total simulation time used.

On p. 4 210 we mentioned that in this study 'aerosol dynamics” means evaporation/activation of aerosol particles.

Also reformulated:

One should also mention, that in the scope of this model we neglect collisions and coalescence of aerosol particles (crucial in creation of rain drops) because of the short total simulation time. Also, due to the short simulation time the droplets are too small for an effective coalescence process.

• p. 7, l. 380ff: “We find that the number : : : linearly depends: : :.” Please check the English and be careful when using three simulations only. Especially Table 3 (p. 5) contradicts. Better skip that sentence or perform more simulations in more narrow Ntot steps.

Reformulated

We find that that at t= 3 s the number of activated particles proportional to the total number, whereas the change of Ntot by a factor of hundred increases LWC by approximately 40 % (Table3}).

• p. 7, l. 400f: “We find that the vertical motion of air is decelerated because of aerosol dynamics.” This contradicts to the statement of neglecting aerosol dynamics (condensation and coagulation) during the period of simulation (p. 7, l. 353)! Please check.

We assume that aerosol dynamics includes condensation and evaporation (it was mentioned in the text). Coalescence and collisions are neglected. Indeed, vertical motion is accelerated because of condensation: the temperature inside the domain decreases, the difference between temperatures inside and outside the domain increases and the buoyancy force also increases.

• p. 8, l. 405ff: You explain the air temperature change driven by the condensation of water vapor onto the aerosol particles and the release of latent heat. But since the aerosol particles are rather huge size shouldn't matter and the condensation should occur independent on the number if any particle number and time are available. But the change differs notably between 55 and 550 cm□3 and I can only think of not sufficient time for condensation.

We added a new figure to show time dependence of supersaturation averaged over domain.
In this Fig. we present the supersaturation averaged over domain for {\it cases 1, 2, 3 and 4} (see Table 1) and analyze the phase relaxation time of supersaturation t_phase for different values of initial supersaturation and total number of particles. Analyzing numerical results we get the relaxation time of about 0.77 s (case 2), 0.17 s (case 3), and 0.6 s (case 4). In case 1 the phase relaxation time is larger than time of simulations. We fit the corresponding curve by exp(-t/tau) and get t_phase=4 s.

• p. 8, l. 436f. The information on the model sizes is very nice but would be best to include it earlier in the methods section for a better understanding on set-up and interpretation of results.

done

• p. 8, l. 450ff: Very nice indeed. But simulating a 10x10x10 cm³ volume this would cause dramatic horizontal and vertical gradients and motion. Is this still applicable by the present method including the problematic areas along the edges of the finite volume?

To analyze the effect of aerosol and droplets on turbulence a small volume with supersaturation of 10% was considered. Under such extreme conditions, condensation was the dominant process. The results cannot be linearly extended to bigger cloud volumes but should be considered as relevant for a small cloud parcel with extreme supersaturation due to turbulent mixing of the water vapor and temperature field.

• p. 8, end: Very nice and interesting results indeed. I would recommend a short statement to potential implications for cloud simulations and weather prognosis. This would definitely increase the range of potential readers, for which the area is highly relevant.

At the end of the manuscript we added

To analyze the effect of aerosol dynamics on the turbulent kinetic energy and on vertical velocity we take the maximal value of $N_{tot}=5550$ cm^{-3}. We conclude that the presence of aerosol has an effect on vertical motion and in our case (when the temperature inside the domain is larger than in environment) tends to enlarge upward velocity. We conclude that aerosols quite strongly influences the dynamics in the cloud area. Such effect of aerosols can be crucial also for large scales usually studied with Large Eddy Simulation (LES) and the LES parametrization can be improved with Direct Numerical Simulations.

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A DNS study of aerosol and small-scale cloud turbulence interaction

N. Babkovskaia[1], U. Rannik[1], V. Phillips[2], H. Siebert[3], B. Wehner[3], and M. Boy[1]

[1]University of Helsinki, Department of Physics, Helsinki, Finland
[2]Lund University, Department of Physical Geography and Ecosystems Science, Sweden
[3]Leibniz Institute for Tropospheric Research, Leipzig, Germany

Correspondence to: N. Babkovskaia
(NBabkovskaia@gmail.com)

Abstract. The purpose of this study is to investigate the interaction between small-scale turbulence and aerosol and cloud microphysical properties using Direct Numerical Simulations (DNS). We consider the domain located at the height of about 2000 m from the sea level, experiencing transient high supersaturation due to atmospheric fluctuations of temperature and humidity. **To study the effect of total number of particles (N_{tot}) on air temperature, activation and supersaturation we vary N_{tot}. To investigate the effect of aerosol dynamics on small-scale turbulence and vertical air motion we vary the intensity of turbulent fluctuations and the buoyant force. We find that even small amount of aerosol particles (55.5 cm^{-3}) and, therefore, small droplet number concentration strongly affects the air temperature due to release of latent heat. The system comes to an equilibrium faster and the relative number of activated particles appears to be smaller for larger N_{tot}. We conclude that aerosol particles strongly affect the air motion. In a case of updraft coursed by buoyant force the presence of aerosol particles results in acceleration of air motion in vertical direction and increase of turbulent fluctuations.**

1 Introduction

Interaction of atmospheric turbulence with aerosol and cloud formation processes has been studied extensively. Due to non-linearity of particle formation and other aerosol dynamical processes, the fluctuations of temperature and relative humidity can have strong effect on aerosol formation. Large scale fluctuations of atmospheric properties, which occur for example in the atmospheric boundary layer, can be the drivers for initiation of particle formation (Easter et al., 1994). Mixing of air with different properties, including temperature and relative humidity, have been shown to enhance atmospheric nucleation significantly (Nilsson and Kulmala, 1998). More specifically, the atmospheric waves can increase the nucleation rate several orders of magnitude and affect also the size spectrum of the particles (Nilsson et al., 2000). Also the activation of atmospheric particles in cloud areas is affected by the fluctuation of supersaturation. Some droplets were shown to grow also in undersaturated conditions due to fluctuations and the bi-modal particle size distribution could be observed after initially unimodal particle population experienced fluctuating supersaturation (Kulmala et al., 1997).

In-cloud turbulence has been shown to intensify cloud-microphysical processes determining cloud properties (Benmoshe and Khain, 2014). Similarly, aerosol loadings also influence the numbers and sizes of cloud-particles, thereby influencing precipitation, cloud extent and hence the climate (Forster, 2007). Both turbulence and aerosol particles can influence the chemical reactions in the atmosphere providing surfaces for aqueous phase chemistry and promoting uptake of gaseous species. This can have repercussions for chemical reactions in the air. Yet both environmental factors, namely aerosol composition and turbulence, have been usually considered as separate influences.

The large-scale atmospheric turbulence is well known to affect aerosol processes significantly. In turn, aerosol properties may influence the buoyancy and turbulence. A motivation for a potential aerosol-turbulence interaction is that the most advanced cloud-microphysical models can now resolve the cloud edges where mixing occurs and where spatial gradients are great in turbulence and concentrations of hydrometeors, including aerosols. Consequently, by including a more complete set of such interactions in the models, the simulation of aerosols , turbulence and microphysics may be improved.

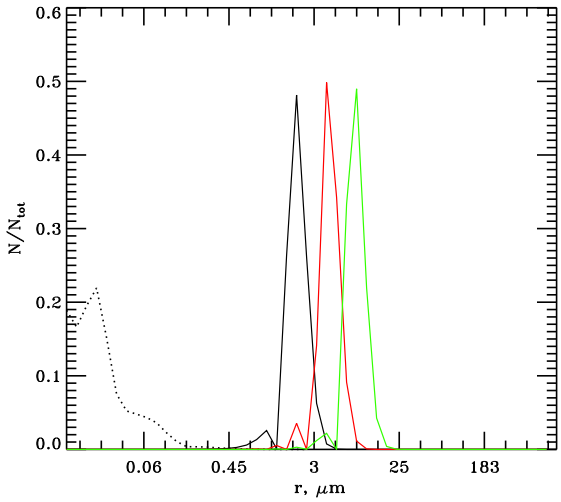

Fig. 1. Relative distribution of particles averaged over the domain at t=0 s (*black dotted curve*) and at $t = 3$ s (*solid curve*) for the cases of $N_{\rm tot} = 55.5$ cm^{-3} (*green solid curve*), $N_{\rm tot} = 555$ cm^{-3} (*red solid curve*), $N_{\rm tot} = 5550$ cm^{-3} (*black solid curve*). Dotted curve corresponds to the observed distribution of aerosol; solid and dashed curves are distributions of droplets.

case 1	$N_{tot} = 55.5$ cm^{-3}	$S_{av} = 10.3$ %
case 2	$N_{tot} = 555$ cm^{-3}	$S_{av} = 10.3$ %
case 3	$N_{tot} = 5550$ cm^{-3}	$S_{av} = 10.3$ %
case 4	$N_{tot} = 5550$ cm^{-3}	$S_{av} = 0.6$ %
case 5	no particles	

Table 1. The summary of key parameters for studying the effect of total number of particles on air temperature, supersaturation and activation. S_{av} is initial supersaturation averaged over the domain.

	equilibrium	*non-equilibrium*
low intensive turbulence	$T_0 = 285.4$ K $f_0 = 10$	$T_0 = 283.5$ K $f_0 = 10$
high intensive turbulence	$T_0 = 285.4$ K $f_0 = 100$	$T_0 = 283.5$ K $f_0 = 100$

Table 2. The summary of key parameters for studying the effect of aerosol on turbulence.

The main goal of this direct numerical simulation (DNS) study is to investigate interaction between small-scale turbulence and aerosol and cloud microphysical properties. The chosen DNS domain is realistic for a small volume at cloud edge where turbulent mixing is a dominate feature (Katzwinkel et al., 2014) and can create fluctuations in temperature as well as humidity. Such conditions however appear transient because presence of atmospheric aerosols leads to depletion of supersaturation via condensation and droplet activation.

We use the high order public domain finite difference PENCIL Code for compressible hydrodynamic flows. The code is highly modular and comes with a large selection of physics modules. It is widely documented in the literature (Dobler et al., 2006; Pencil Code, 2001, and references therein). The chemistry module is responsible for the detailed description of the necessary quantities in a case of complicated chemical composition, such as diffusion coefficients, thermal conductivity, reaction rates etc. (Babkovskaia et al., 2011). The detailed description of aerosol module can be found in Babkovskaia et al. (2015). The paper is constructed as follows. Section 2 is devoted to the description of the model. Results are presented in Section 3. Section 4 provides the summary of our study.

2 Description of the model

Aerosol

As a starting point for our simulations we consider values typical for observations made in trade wind cumuli. During the CARRIBA project (Siebert et al. 2013, hereafter called SI13) total aerosol number concentrations in the marine subcloud layer up to a few 100 cm^{-3} have been observed (Fig 6f in SI13). Although it was argued that the highest values are due to local biomass burning we consider a concentration of 550 cm^{-3} as typical and 55 and 5550 cm^{-3} as two extreme values for sensitivity test. The initial normalized aerosol number size distribution as shown as a dotted line in Fig. 1 compares well with the shape of the observed distribution shown as red line in Fig. 8 of SI13.

We assume a soluble aerosol (NaCl) which will dilute inside droplet. We take 50 size bins logarithmically distributed in the range [10 nm, 1000 μm]. As an initial distribution of particles we take the observational data at the sea level and assume that it is the same everywhere in the domain (see the distribution in Fig. 1). To analyze the effect of total number of aerosol particles, N_{tot}, on the structure and properties of turbulent motion we consider the following cases: $N_{tot} = 55.5$ cm^{-3} and initial supersaturation averaged over domain $S_{av} = 10.3$ % (*case 1*); $N_{tot} = 555$ cm^{-3} and $S_{av} = 10.3$ % (observed data, *case 2*); $N_{tot} = 5550$ cm^{-3} and $S_{av} = 10.3$ % (*case 3*); $N_{tot} = 5550$ cm^{-3} and $S_{av} = 0.6$ % (*case 4*); and no particles (*case 5*), see Table 1.

Air composition

We assume the following air composition O_2 + H_2O + N_2, where nitrogen mass fraction is taken to be $Y_{N_2} = 70\%$. The

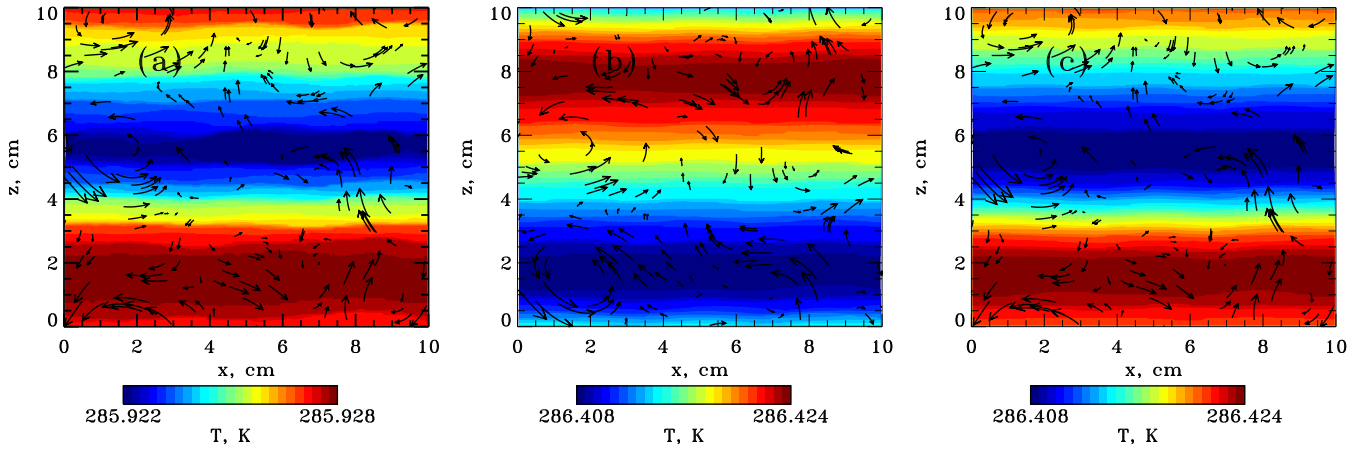

Fig. 2. Temperature distribution at $t = 3$ s in *case 1* (a), *case 2* (b) and *case 3* (c), see Table 1. *Non-equilibrium case with low intensive turbulence* is considered (see Table 2). Velocity vector is shown by arrows (averaged vertical velocity is subtracted).

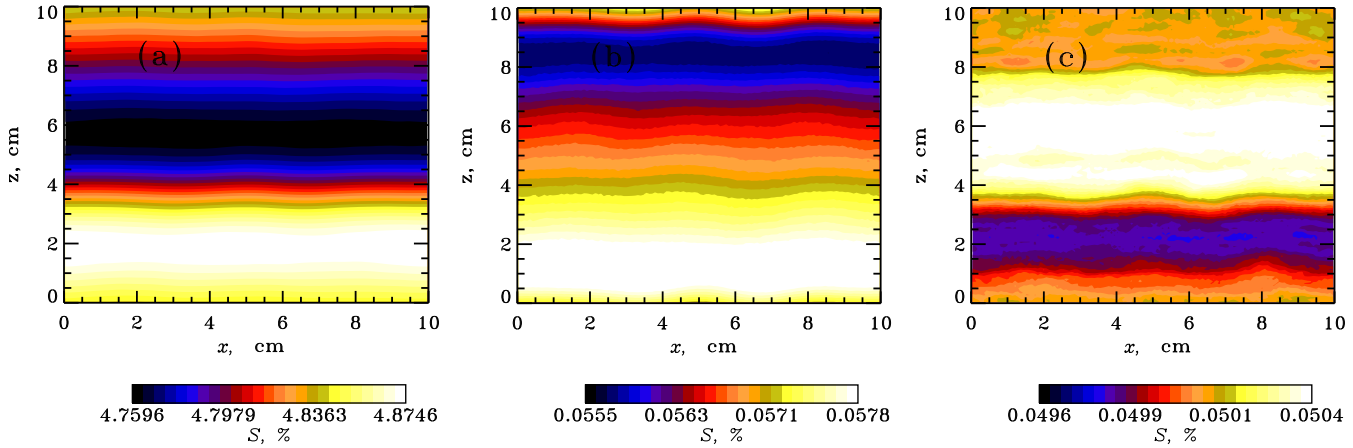

Fig. 3. Distribution of supersaturation at $t = 3$ s in *case 1* (a), *case 2* (b) and *case 3* (c), see Table 1. *Non-equilibrium case with low intensive turbulence* is considered (see Table 2).

observations provide us the absolute humidity to set the initial value for mass fraction of water vapor, Y_{H_2O}. Oxygen mass fraction is recalculated from the normalization conditions, i.e. $Y_{O_2} + Y_{H_2O} + Y_{N_2} = 1$, where Y_{O_2} is oxygen mass fraction.

Initial conditions

This model represents the 3D fluid flow on the microscale inside a volume of 10 cm x 10 cm x 10 cm, just inside the cloud in the mid-troposphere. Based on data of CARRIBA observations typical for the upper parts of clouds / cloud edges at a height of 2000 m, we set the initial conditions for air temperature ($T_0 = 285.4$ **K) and water vapor mixing ratio** ($q_0 = 0.0124$)**. The small vertical gradients of temperature and water content are also based on the CARRIBA measurements: the total difference between values of air temperature and water vapor mixing ratio at the upper and lower edges of the domain are** $\Delta T = 0.001$ **K and** $\Delta q = 4 \times 10^{-5}$**, correspondingly.**

Observed temperature and absolute humidity result in peak value of supersaturation (S) of up to 0.1 (e.g. 10 %). Such values of S are extremely high considering only an adiabatic lifting of a cloudy air parcel. This "quasi-steady state" supersaturation depends mainly on vertical updraft velocity, droplet number concentration, and mean droplet diameter and appears to be on the order of a few tenths of percent for vertical updrafts of about 1 m s^{-1}. However, it is well known that atmospheric clouds are mainly non-adiabatic due to turbulent mixing, and a few percent supersaturation are realistic for higher updraft velocities (e.g., Korolev and Mazin, 2003). This is particular true at cloud edges where entrainment of unsaturated air into the cloud results in strong mixing and fluctuations of the water vapor and temperature field. It strongly depends on the correlation between these two ther-

modynamic fields whether strong fluctuations can result in high fluctuations of supersaturation or not. Until now we are not aware of any observations of this correlation in clouds. Based on theoretical arguments and observations in the convective boundary layer below clouds Kulmala et al. (1997) provided some convincing arguments that at the cloud base high fluctuations of supersaturation on the order of up to several percent can exist. It is also well known that turbulence in high Reynolds number flows (typical in convective clouds) is highly intermittent (Siebert et al., 2010). Shaw (2000) argued that under such conditions long-living vortex tubes could produce small areas with decreased droplet number concentrations and, therefore, high supersaturation resulting in secondary activation. Summarizing, there are good arguments that supersaturation fluctuations of a few percent can be generated without strong updrafts.

On the other hand, the supersaturation excess would be eliminated by condensation onto droplets and quasi-steady state supersaturation would be restored (Korolev and Mazin, 2003). Therefore, the key question about the temporal time scale is under consideration now. The ratio of two time scales is important here: the phase relaxation time which describes how fast the supersaturation can react on the new thermodynamic condition and the turbulent mixing time scale which describes how fast turbulence can mix a certain volume (eddy with the length scale l). **If the phase relaxation time is smaller than the turbulent mixing time then the actual supersaturation will tend to the quasi steady-state solution. However, for scales where the turbulent mixing is faster we expect strong supersaturation fluctuations to "survive".**

Let us now assume a small eddy of size $l = 1$ m and a local energy dissipation rate of $\epsilon = 0.1\mathrm{m}^2\mathrm{s}^{-3}$. These dissipation values are typical peak values for cumulus clouds on that small scales. The highest dissipation can be found at Kolmogorov size (see discussion in Siebert et al., 2006, 2010). The eddy turn-over time is $\tau_{eddy} = (l^2/\epsilon)^{1/3} \approx 2$ s. If we now take a phase relaxation time of the order of one second which is typical for cumulus clouds (see again Korolev and Mazin (2003)) we see that these two time scales are of the same order. Therefore, we conclude that on scales below one meter, strong supersaturation fluctuations can exist and the quasi-steady state solution should be considered as a mean value with superimposed fluctuations with amplitudes up to several percent. This argumentation partly follows the discussion in Section 8e by Korolev and Mazin (2003).

Kulmala et al. (1997) estimated standard deviations of supersaturation of up to 5 % based on aircraft observations at cloud level but outside the clouds. Ditas et al. (2012) observed supersaturation fluctuations in a field of stratocumulus clouds and estimated from highly collocated temperature and water vapor observations peak-to-peak values of up to 1.5 %, which is much higher compared to the quasi-steady-state solution. It is straightforward to assume that higher S values can be expected for parts of (shallow) cumulus clouds. Thus, we argue that for our small modeled volume a transient supersaturation of 10 % can be realistic for specific mixing event.

The pressure, p, is assumed to be constant everywhere in the domain and it is also based on measurements. The air density, ρ, is calculated from the equation of state $p = \rho RT/m$, where R is gas universal constant, m is air molar mass. The initial velocity is taken to be zero.

To generate the initial turbulent field we make first 100 iterations without evaporation/activation of aerosol particles (further "aerosol dynamics"), including randomly directed external forces (see next section). After that the external forces are set to zero, whereas the particles start to evolve. Thus, the turbulence is decaying for the analyzed time. The maximal time step allowed by the Courant condition for convergence is $\Delta t_c = 10^{-6}$ s (Courant time step). Since Δt_c is much larger than the time step needed to move the smallest particle to the neighbor size bin ($\Delta t_a = 2 \times 10^{-7}$ s), at every Courant time step we make 5 sub steps and calculate the particle evolution equation only.

Boundary conditions

In all three directions we set periodic boundary conditions for all variables, including the number density function. It means, that at every time step Δt_c the number of particles appearing on the bottom/left boundaries is equal to the number of particles disappearing through the top/right boundaries. While the periodic boundary conditions modify the initial temperature stratification, they allow us to consider this domain as an isolated volume, i.e. the total mass, energy and number of particles in the domain does not change with time. In turn, it makes possible to compare the results of simulations, varying the key parameters of the model and to carry out the detailed quantitative analysis of the interaction between aerosol and turbulence.

Basic equations

The detailed description of the main equations is presented by Babkovskaia et al. (2015). We consider the standard compressible Navier-Stokes system including equation for conservation of mass, momentum, energy and chemical species. The momentum of air is transported due to viscous force. To describe the gravitational effect we add the buoyant force B to vertical component of momentum equation as

$$B = g\left[\frac{T - T_0}{T_0} + \epsilon(Y_{H_2O} - q_0) - q_c\right], \quad (1)$$

where $g = 9.81$ m s^{-2} is the acceleration of gravity, q_0 is the reference water vapor mixing ratio, $\epsilon + 1 = R_v/R_d$ is the ratio of the gas constant for water vapor and dry air, and q_c is the cloud water mixing ratio (Andrejczuk et al., 2004; Babkovskaia et al., 2015). Note, that the

buoyancy force is applied to the air vertical acceleration/deceleration. The air temperature can evolve due to thermal diffusion, viscous heating, adiabatic contraction/expansion and latent heating/cooling caused by condensation/evaporation on the droplet surface. Coefficient of thermal conductivity and kinematic viscosity are calculated for a mixture of three air species.

We consider evolution of aerosol number density function happening because of evaporation/condensation of aerosol particles. There is energy exchange between particles and ambient gas due to release/absorb of latent heat caused by condensation/evaporation on the droplet surface. The motion of the particles is determined exclusively by their involvement in the motion of surroundings. In the present study we modify water vapor pressure over a droplet of radius r as (Seinfeld and Pandis, 2006; Sorjamaa and Laaksonen, 2007)

$$p_{vs} = p_0 \exp\left(\frac{A}{2r} - \frac{0.1 d_w}{2(r - r_0)}\right) \quad (2)$$

where p_0 is water vapor pressure over a flat surface at the same temperature, $A = 0.66/T$ (in μm), where $r_0 = 10$ nm is the radius of the droplet core, $d_w = 0.3$ nm is the size of water molecule.

For generation of the initial turbulence the external forcing f is used in a form

$$f(\mathbf{x}, t) = Re\{N f_k(t) \exp[i\mathbf{k}(t) \cdot \mathbf{x} + i\phi(t)]\}, \quad (3)$$

where $\mathbf{x}$ is the position vector. The wave vector $k(t)$ and the random phase $-\pi < \phi(t) \leq \pi$ change at every time step; $N = f_0 c_s (|k| c_s / \Delta t_c)^{1/2}$ is the normalization factor, c_s is the sound speed, f_0 is a non-dimensional forcing amplitude; $f_k = (\mathbf{k} \times \mathbf{e}) / \sqrt{\mathbf{k}^2 - (\mathbf{k} \cdot \mathbf{e})^2}$, where $\mathbf{e}$ is an arbitrary unit vector that is real and not aligned with $\mathbf{k}$ (see detail in Pencil Code (2001)).

To study the effect of aerosol on the turbulence we consider two initial turbulent fields, taking different non-dimensional forcing amplitudes as $f_0 = 10$ (*low intensive turbulence*) and $f_0 = 100$ (*high intensive turbulence*). Also, we compare the effect of the aerosol in the case when the domain is in equilibrium with environment (*equilibrium* case) and when the temperatures inside and outside the domain are different (*non-equilibrium* case), taking the environment temperature $T_0 = 285.4$ K for *equilibrium case* and $T_0 = 283.5$ K for *non-equilibrium case* . The summary of four considered cases is in Table 2. To study the effect of total number of particles on activation, air temperature and supersaturation we consider *non-equilibrium case* with *low intensive turbulence*.

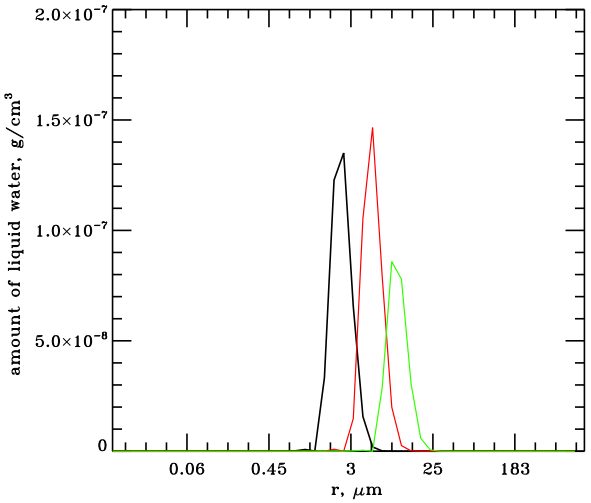

Fig. 4. Amount of liquid water accumulated in particles with corresponding radius r at $t = 3$ s (*solid curve*) for the cases of $N_{\rm tot} = 55.5$ cm^{-3} (*green solid curve*), $N_{\rm tot} = 555$ cm^{-3} (*red solid curve*), $N_{\rm tot} = 5550$ cm^{-3} (*black solid curve*).

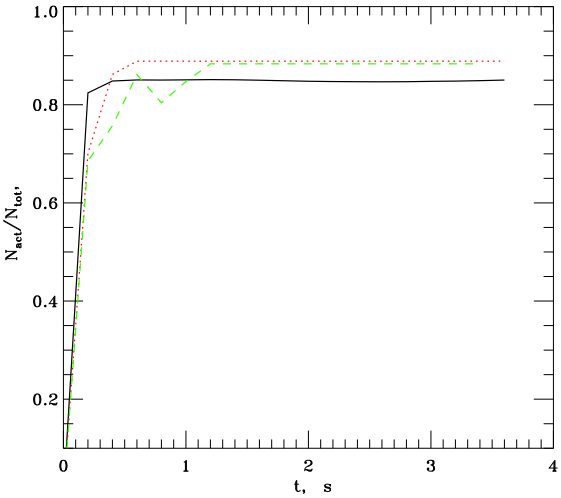

Fig. 5. Relative number of activated particles as a function of time $N_{\rm tot} = 55.5$ cm^{-3} (*green curve*), $N_{\rm tot} = 555$ cm^{-3} (*red curve*), $N_{\rm tot} = 5550$ cm^{-3} (*black curve*).

	case 1	*case 2*	*case 3*	*case 4*
$\overline{S_{init}}$, %	10.3	10.3	10.3	0.6
N_{tot},cm^{-3}	55.5	555	5550	5550
N_{act}, cm^{-3}	49	493	4700	395
LWC, g/m^3	0.23	0.369	0.376	0.0377
Δ T, K	0.52	1.0	1.0	0.05
Δ S$_{max}$, %	-5.42	-10.11	-10.47	-0.69
Δ S$_{min}$, %	-5.66	-10.12	-10.48	-0.37
τ_{phase}, s	4	0.77	0.17	0.6

Table 3. Initial value of supersaturation averaged over the domain $\overline{S_{init}}$, total number of particles (N_{tot}), number of activated particles (N_{act}) at $t = 3$ s, liquid water content (LWC) at $t = 3$ s, change in temperature between start and end of simulation (Δ T), change in percentage maximal supersaturation between start and end of simulation (Δ S$_{max}$), change in percentage minimal supersaturation between start and end of simulation (Δ S$_{min}$), the phase relaxation time of supersaturation τ_{phase} at $t = 3$ s for considered *cases 1, 2, 3, 4* (see Table 1). The phase relaxation time τ_{phase} is the numerically predicted value in *cases 2, 3, 4*. In *case 1* the phase relaxation time is obtained from Eq. 6. No subsaturation is predicted anywhere in the domain in all cases. *Non-equilibrium* case with *low intensive turbulence* is considered (see Table 2).

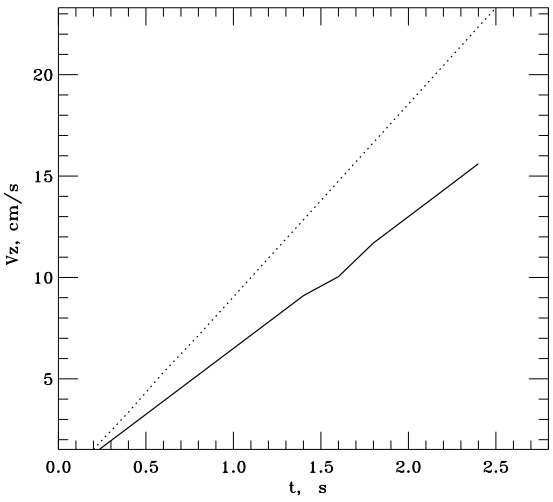

Fig. 6. Averaged vertical velocity as a function of time with aerosol for $N_{\rm tot} = 5500$ cm^{-3} (*dotted curve, case 3*); and without aerosol (*solid curve, case 5*). *Non-equilibrium* case with *low intensive turbulence* is considered (see Table 3).

3 Results

Effect of total number of particles on air temperature and supersaturation distributions

In Fig. 2 we present the air temperature averaged in y-direction when the aerosol dynamic is included. We find that in *case 3* (see Fig. 2, c) after 3 s the difference between the absolute value of maximal and minimal temperatures in the domain is about 0.02 K, whereas without aerosol (*case 5*, not shown) after 3 s the temperature difference is still about 0.001 K. Moreover, the temperature increases by about 1 K everywhere in the domain because of condensation of water vapor on aerosol particles.

Changing of temperature distribution with time is mostly attributable to the periodic boundary conditions: the coldest layers are moving from the bottom to the middle of the domain. However, comparing (a), (b) and (c) panels in Figs. 2 and Fig. 3 we note that the positions of the coldest layers in different panels are different. The coldest layers in case of $N_{\rm tot} = 55$ cm^{-3} coincide with the layers where supersaturation is minimal (see panel a), whereas for other two cases, $N_{\rm tot} = 555$ cm^{-3} (b) and 5550 cm^{-3} (c), the coldest layers correspond to the positions where supersaturation achieves maximum. It happens because of the different effect of aerosol dynamics in dependence on $N_{\rm tot}$. The small amount of aerosol particles (*case 1*) does not make any substantial effect on the temperature distribution, i.e. both temperature and supersaturation are identically shifting with time in vertical direction. In turn, in *cases 2* and *3* aerosol dynamics crucially changes the temperature distribution. One can see in Figs. 2, 3 that layers with larger temperature correspond to layers with smaller supersaturation. It can be interpreted as follows. More intensive condensation occurs in initially warmer layers because supersaturation is larger there and respectively the temperature increases faster in these layers. In turn, since supersaturation exponentially depends on temperature, $S \propto \exp(-T)$, at some moment S appears to be smaller in warmer than cooler layers. Thus, equilibrium supersaturation is higher in the layers with temperature minimum (and vice versa). Also, we find that in *case 1* at $t = 3$ s the supersaturation is about 5 % and the aerosol is still activating, whereas in *case 3* for the first 3 s the supersaturation almost drops to zero and the system is coming to an equilibrium.

Additionally, we investigate how fast the system with initially high value of supersaturation ($S = 10$ %) comes to an equilibrium ($S \simeq 0$ %) in dependence on different total numbers of droplets to answer the question how $N_{\rm tot}$ affects the phase relaxation time. Analyzing Fig. 3 we find that in a case of $N_{tot} = 55$ cm^{-3} it takes more than 3 s for the system to come to equilibrium, i.e. phase relaxation time appears to be larger than theoretically estimated turbulent mixing time ($\tau_{eddy} = 2$ s) (see discussion in section 2). Thus, strong supersaturation fluctuations can "survive" longer if the total number of droplets is small.

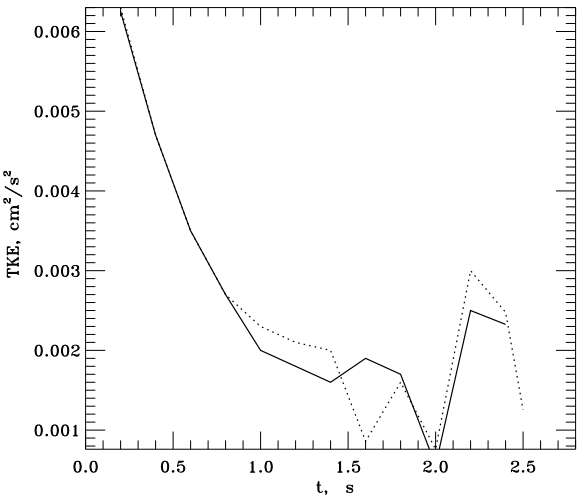

Fig. 7. The dependence of the average turbulent kinetic energy on time with aerosol for $N_{\rm tot} = 5500$ cm^{-3} (*dotted curve, case 3*); and without aerosol (*solid curve, case 5*). The other parameters correspond to *non-equilibrium* case with *low intensive turbulence* (see Table 2).

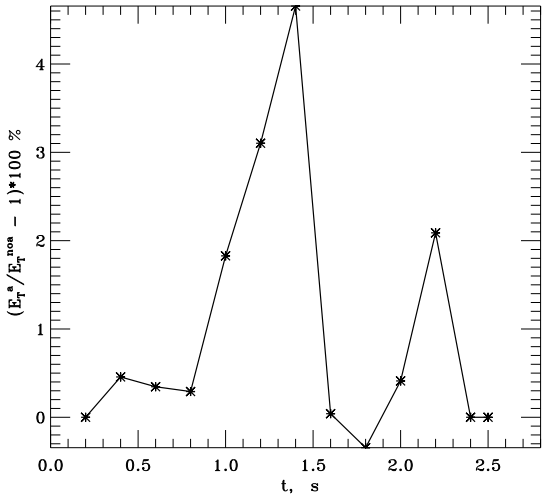

Fig. 8. The difference between turbulent kinetic energy averaged over time period t in a case without aerosol (E_T^{noa}) and with aerosols (E_T^a), where $E_T(t) = 1/t \int_0^t TKE(t')dt'$. All other parameters are the same as in Fig.7.

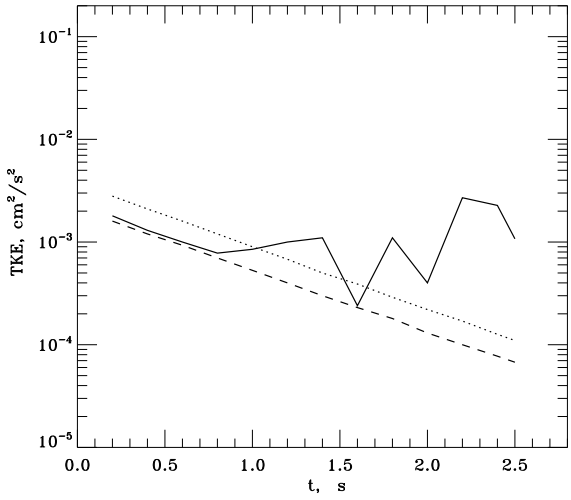

Fig. 9. The dependence of the x-component (*dotted curve*), y-component (*dashed curve*) and z-component (*solid curve*) of the averaged kinetic energy on time with aerosol for $N_{\rm tot} = 5500$ cm^{-3}.

Effect of total number of particles on activation

In this study we consider that all particles with radius larger than $r_{cr} = 1.75$ μm are activated. This value is somewhat arbitrary but the results of our study were not sensitive to the choice of r_{cr} provided that $r_{cr} \leq 1.75$ μm. In Fig. 1 we present the normalized particle distribution at $t = 0$ s and $t = 3$ s. We find that the smaller the total number the larger particles are produced for the similar time period. Comparing the supersaturation at $t = 3$ s in these three cases (Fig. 3) one can see that in the case of the largest N_{tot} the supersaturation appears to be close to zero and particles stop to grow, while for the smaller N_{tot} the supersaturation is about 4.8 % and particles continue growing. Therefore, the system is coming to an equilibrium (i.e. $S \simeq 0$) faster and aerosol particles cease to grow earlier for the larger N_{tot}. Indeed, the phase relaxation time can be estimated as

$$\tau_{phase} = (a_2 N_{tot} \overline{r})^{-1}, \quad (4)$$

where $\overline{r}$ is the mean droplet radius and $a_2 = 3.04\ 10^{-4}$ m^2/s. **The steady-state supersaturation can be written as $S_{qs} \propto a_1 w \tau_{phase}$, where a_1 is a parameter including thermodynamic parameters and being almost constant, w is vertical velocity. Thus, it becomes clear that for larger N_{tot} the system comes to an equilibrium faster. We show that aerosol particles can grow and possibly achieve the size of the rain drop only in the case of small total number of particles. This is consistent with the fact that in natural clouds, deep ascent is needed to drive the prolonged con-**

densational growth of drops in order to form rain (e.g. Rogers and Yau 1989, a short course in cloud physics). In real clouds, the ascent is the source of larger-scale supersaturation. One should also mention, that in the scope of this model we neglect collisions and coalescence of aerosol particles (crucial in creation of rain drops) because of the short total simulation time. Also, due to the short simulation time the droplets are too small for an effective coalescence process.

In addition, in Fig. 4 we analyze the amount of liquid water, $LW(r) = (4/3)\pi r^3 \rho_w N(r)$, where $N(r)$ is the number of particles with radius r, and ρ_w is liquid water density. In Fig. 5 we show the fraction of activated particles averaged over the domain as a function of time in *cases 1, 2, 3*. In Table 3 we collect the most important quantities, such as initial value of supersaturation ($\overline{S_{init}}$), number of activated particles (N_{act}) at $t = 3$ s, liquid water content $LWC = \int LW(r)/\Delta r \, dr$ (where Δr is a size of corresponding bin), change in temperature between start and end of simulation (Δ T), change in percentage maximal supersaturation between start and end of simulation (Δ S_{max}), change in percentage minimal supersaturation between start and end of simulation (Δ S_{min}), the phase relaxation time of supersaturation τ_{phase} for *cases 1, 2, 3, 4* (see Table 1). Note that no subsaturation is predicted anywhere in the domain in all considered cases.

We find that while the total number in *case 2* is ten times smaller than in *case 3*, liquid water content is similar in these two cases. On the other hand, *LWC* appears to be smaller in *case 1* than in *cases 2, 3* (see Table 3 and Fig. 4). It happens because the probability of water molecules to catch a particle is much smaller in a case of the smallest particle concentration (*case 1*) than in *cases 2, 3*. In Fig. 5 one can see that in *case 1* and *case 2* the number of activated particles does not grow after 1.2 s and 0.6 s, correspondingly, and in *case 3* it happens after 0.5 s. Since in *case 3* the equilibrium is achieved earlier than in *cases 1* and *2*, the maximum of final particle distribution in *case 3* is shifted (see Fig. 1) and the final relative number of activated particles appears to be smaller than in *cases 1* and *2*. We find that at t= 3 s the number of activated particles is proportional to the total number, whereas the change of N_{tot} by a factor of hundred increases *LWC* by approximately 40 % (Table 3).

Effect of aerosol on the turbulent motion

We analyze the effect of aerosol on the turbulent motion, taking $N_{tot} = 5500$ cm^{-3} (*case 3* in Table 1). First, we consider the *equilibrium case*, taking $T_0 = 285.4$ K and $q_0 = 0.0124$ in Eq. 1. In that case the turbulent field appears to be isotropic. Next, we decrease $T_0 = 283.5$ K and study the developing of turbulence in *non-equilibrium case*. In that case the intensive vertical motion is generated due to buoyant force. Note, that the model domain is not vertically displaced during the simulation time but the vertical motion is generated within domain. Also, we vary parameter f_0 in Eq. 3 to compare the developing of *low intensive* ($f_0 = 10$) and *high intensive* ($f_0 = 100$) turbulence in both *equilibrium* and *non-equilibrium* cases. The summary of parameters is presented in Table 2. Finally, we compare the results of simulations with and without particles in all four cases described above.

We find that the vertical motion of air is accelerated because of aerosol dynamics. Also, in Fig. 8 we show the time averaged turbulent kinetic energy, $E_T(t) = 1/t \int_0^t TKE(t')dt'$, as a function of time in the case with aerosol particles and without them. We conclude that turbulent kinetic energy increases because of presence of aerosols. We interpret these results as follows. The air temperature increases because of release of latent heat caused by condensation onto droplets, and therefore, the difference between temperatures inside and outside the domain is enlarged. It results in increasing of buoyant force and accelerating of air motion in vertical direction.

Also, we find that acceleration does not depend on intensity of turbulent fluctuations, i.e. acceleration in vertical direction is the same in *low intensive turbulence* and in *high intensive turbulence* cases. Moreover, turbulent fluctuations grow because of presence of aerosol particles in all four considered cases. The dependences of vertical velocity and turbulent kinetic energy (TKE) averaged over the domain as a function of time for *non-equilibrium* case with *low intensive turbulence* are presented in Figs. 6 and 7, correspondingly.

Finally, we find that there is a strong variation of TKE with time (see Fig. 7). To interpret this fact we plot *x*- ,*y*- and *z*- components of TKE in Fig. 9 and find strong time variations only in *z*-component of TKE (*x*- and *y*- components are smoothly decreasing with time). We conclude that it results from fluctuations of temperature caused by aerosol dynamics, and therefore, changes of buoyant force with time, which result in perturbation of vertical motion.

Effect of initial supersaturation on activation of aerosol particles

To illustrate the effect of initial supersaturation on activation of aerosol particles we modify the initial distribution of absolute humidity in *case 3* decreasing it by 10 %. In that case the initial supersaturation averaged over domain becomes 0.6 %. To study the effect of initial supersaturation on activation of aerosol particles in Fig. 10 we analyze count median diameter (CMD) taking

$$\mathrm{CMD} = (D_1^{n_1} D_2^{n_2} D_3^{n_2} ... D_{N_{tot}}^{n_{N_{tot}}})^{1/N_{tot}}, \quad (5)$$

where D_i is particle size of the i^{th} bin, n_i is number of particles having a size D_i. Dependences of CMD on time in *cases 1, 2, 3, 4* are presented in Fig. 10. Comparing *cases 3* and *4* we find that in both cases the system comes to equilibrium at $t \simeq 0.6$ s. In other words, the typical time it takes for droplets to cease growing does not depend

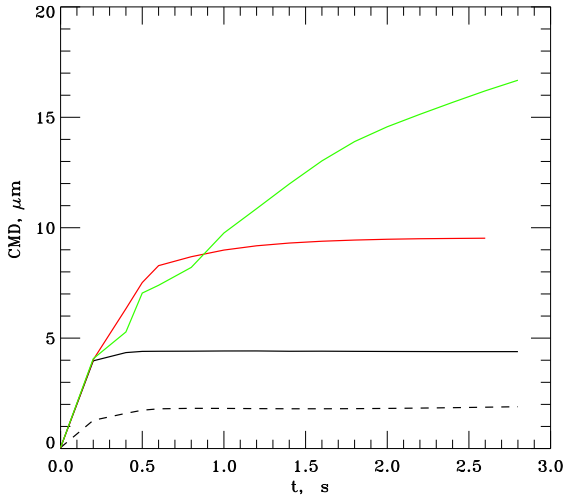

Fig. 10. The dependence of count mean diameter (CMD) on time for *case 1* **(***green solid curve***),** *case 2* **(***red solid curve***),** *case 3* **(***black solid curve***),** *case 4* **(***black dashed curve***).**

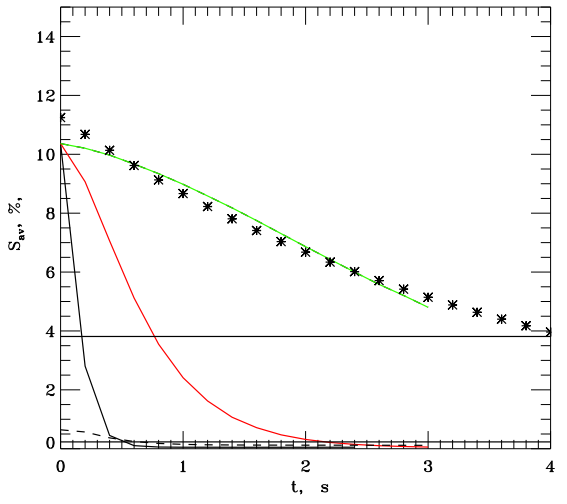

Fig. 11. The dependence of supersaturation averaged over the calculation domain on time for *case 1* **(***green solid curve***),** *case 2* **(***red solid curve***),** *case 3* **(***black solid curve***),** *case 4* **(***black dashed curve***). Horizontal lines show the corresponding values of $\overline{S_{init}}/e$. Asterisk show the extrapolation function in** *case 1* **(see Eq. 6).**

on initial supersaturation. On the other hand, a value of CMD in equilibrium crucially depends on supersaturation and it appears to be two times larger in *case 3* **than in** *case 4***.**

Additionally, in Fig. 11 we present the supersaturation averaged over domain for *cases 1, 2, 3, 4* **(see Table 1) and analyze the phase relaxation time of supersaturation (τ_{phase}) for different values of initial supersaturation and total number of particles. Analyzing numerical results we get the phase relaxation time of about 0.77 s (***case 2***), 0.17 s (***case 3***) and 0.6 s (***case 4***). Using Eq. 4 and count median diameter (see Fig. 10) we estimate the phase relaxation time of 1.23 s (***case 2***), 0.27 s (***case 3***) and 0.63 s (***case 4***). Therefore, we conclude that results of our simulations are in a good agreement with analytical estimations. In** *case 1* **the phase relaxation time is larger than time of simulations. In that case we extrapolate the numerical results with the following function (see asterisk in Fig. 11)**

$$S_{av}(t) = 11.25 \exp\left(-\frac{t}{3.84}\right) \tag{6}$$

and estimate the phase relaxation time of about 4 s. The results are summarized in Table 3. We conclude that τ_{phase} depends both on initial supersaturation and on the total number of particles.

4 Summary

Turbulence, aerosol growth and microphysics of hydrometeors in clouds are intimately coupled. In the present study a new modeling approach was applied so as to quantify this linkage. We study the interaction in the cloud area under transient, high supersaturation conditions, using direct numerical simulations. As the initial conditions we take observational data. To analyze the effect of aerosol and droplets on turbulence a small volume with supersaturation of 10% was considered. Under such extreme conditions, condensation is the dominant process. The results cannot be linearly extended to bigger cloud volumes but should be considered as relevant for a small cloud parcel with extreme supersaturation due to turbulent mixing of the water vapor and temperature field. As an initial distribution of particles we take the data of measurements at the sea level and analyze the droplets activated by the aerosols in the simulations.

To study the effect of total number of particles on activation, air temperature and supersaturation we vary the total number of particles and take the other parameters corresponded to *low intensive turbulence* and *non-equilibrium case*, i.e. when the vertical motion is generated within domain because of buoyancy. We compare the results of simulations with particles and without them. We find that the total number of particles in the domain is crucial for distribution of temperature and for developing of turbulence. Even small amount of aerosol particles (55.5 cm^{-3}) and therefore small

cloud droplet number concentrations increase the air temperature by 1 K because of latent heating caused by condensation onto drops. The system comes to an equilibrium faster for the larger total number of particles. **To illustrate the effect of initial supersaturation on activation of aerosol particles we compare the results of simulations with initial supersaturation averaged over domain of 10.3 % and 0.6 %. We conclude that the typical time it takes for droplets to cease growing does not depend on initial supersaturation. Also, we analyze the phase relaxation time of supersaturation (i.e. time of dropping from initial value to $1/e = 0.368$) for different values of initial supersaturation and total number of particles. We find that the phase relaxation time crucially depends both on the total number of particles and on initial supersaturation.**

To analyze the effect of aerosol dynamics on the turbulent kinetic energy and on vertical velocity we take the maximal value of $N_{tot} = 5550$ cm^{-3}. We conclude that the presence of aerosol has an effect on vertical motion and in our case (when the temperature inside the domain is larger than in environment) tends to enlarge upward velocity. We conclude that aerosols quite strongly influences the dynamics in the cloud area. Such effect of aerosols can be crucial also for large scales usually studied with Large Eddy Simulation (LES) and the LES parametrization can be improved with Direct Numerical Simulations.

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