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

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Abstract. The purpose of this study is to investigate the effect of aerosol dynamics (evaporation/condensation) on atmospheric small-scale turbulence (and vice versa) using direct numerical simulations (DNS). We consider the domain located on 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 aerosol dynamics on the turbulence we vary the total number of particles (N_{tot}). In turn, to investigate the effect of small-scale turbulence on evolution of aerosol particles we vary the intensity of turbulent fluctuations and the buoyant force. We find that even small amount of aerosol particles (55.5 cm$^{-3}$) increases the air temperature by 1 K under supersaturated conditions 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 the presence of aerosol particles results in deceleration of air motion in vertical direction and damping 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, where 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 (Bensoussan and Khain, 2014). Similarly, aerosol loadings also influence the numbers and sizes of cloud-particles, thereby influencing precipitation of formation, 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, a better simulation of aerosols, turbulence and microphysics may be possible.

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 dominant 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 is 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 is the summary.

## 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 sub-cloud 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. 3 in our manuscript 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. 3). To analyse the effect of total number of aerosol particles, $N_{\text{tot}}$, on the structure and properties of turbulent motion we consider the following cases: $N_{\text{tot}} = 55.5$ cm$^{-3}$ (case 1), $N_{\text{tot}} = 555$ cm$^{-3}$ (observed data, case 2), $N_{\text{tot}} = 5550$ cm$^{-3}$ (case 3), and no particles (case 4), see Table 1.

### Air composition

We assume the following air composition O$_2$ + H$_2$O + N$_2$, where nitrogen mass fraction is taken to be $Y_{\text{N$_2$}} = 70\%$. The observations provide us the absolute humidity to set the initial value for mass fraction of water vapour, $Y_{\text{H$_2$O}}$. Oxygen mass fraction is recalculated from the normalization conditions, i.e. $Y_{\text{O$_2$}} + Y_{\text{H$_2$O}} + Y_{\text{N$_2$}} = 1$, where $Y_{\text{O$_2$}}$ is oxygen mass fraction.

### Initial conditions

Based on data of CARRIBA observations typical for the upper parts of clouds/cloud edges in a height of 2000 m, we set the initial conditions for air temperature ($T_0 = 285.4$ K) and water vapour mixing ratio ($q_0 = 0.0124$). The small vertical gradients of temperature ($\Delta T = 0.001$ K) and water content ($\Delta q = 4 \times 10^{-5}$) are also based on the CARRIBA measurements.

Direct observations of supersaturation fluctuations are not possible so far and one has to rely on estimates based on measurements of air temperature and water vapor mixing ratio. The measured temperature and absolute humidity led us to a range of supersaturation ($S$) from 10.17% to 10.54%. Such values of $S$ are extremely high in a case of an adiabatic lifting of a cloudy air parcel. The “quasi-steady state” supersaturation depends mainly on vertical updraft velocity 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 thermodynamic 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)

### Table 1.

| case | $N_{\text{tot}}$ (cm$^{-3}$) |
|------|----------------------------|
| 1    | 55.5                       |
| 2    | 555                        |
| 3    | 5550                       |
| 4    | no particles               |

### Table 2.

| turbulence        | equilibrium $T_0$ | $T_0$ |
|-------------------|------------------|-------|
| low intensive     | $T_0$ = 285.4 K  | 293 K |
| high              | $T_0$ = 285.4 K  | 293 K |

$S = \frac{\text{actual value for mass fraction of water vapour}}{\text{initial value for normalized condition}}$ 

Air composition $O_2 + H_2O + N_2$, nitrogen mass fraction $Y_{N_2} = 70\%$. 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.
provided some convincing arguments that at the cloud base
high fluctuations of supersaturation on the order of up to sev-
eral 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 concen-
trations and, therefore, high supersaturation resulting in sec-
ondary activation. Summarizing, there are good arguments
that supersaturation of a few percent can be generated with-
out strong updrafts.

On the other hand, the supersaturation excess would be elimi-
inated by condensation onto droplets and the usual equi-
librium supersaturation would be restored. 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 tur-
bulent 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 compared to the turbulent
mixing time than the quasi-steady state solution would be
applicable. 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 lo-
cal energy dissipation rate of \( \varepsilon = 0.1 m^2 s^{-3} \). These dissipation
values are typical peak values for cumulus clouds on that
small scales. The highest dissipation can be found at Kol-
mogorov size (see discussion in Siebert et al., 2006, 2010).
The eddy turn-over time is \( \tau_{eddy} = (l^2/\varepsilon)^{1/3} \approx 2 \) 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 or-

Fig. 1. Temperature distribution at \( t = 3 \) s in unequalibrium case for the following cases: \( N_{tot} = 55 \) cm\(^{-3}\) (a), \( N_{tot} = 550 \) cm\(^{-3}\) (b), \( N_{tot} = 5500 \) cm\(^{-3}\) (c). Velocity vector is shown by arrows (averaged vertical velocity is subtracted).

Fig. 2. Distribution of supersaturation at \( t = 3 \) s in the following cases: \( N_{tot} = 55 \) cm\(^{-3}\) (a), \( N_{tot} = 550 \) cm\(^{-3}\) (b), \( N_{tot} = 5500 \) cm\(^{-3}\) (c) in unequalibrium case.
Under. Therefore, we conclude that on scales below one meter, strong supersaturation fluctuations can exist and the quasi-steady state solution is not applicable anymore. This argument partly follows the discussion in Section 8e by Kolev 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 events.

The pressure, $p$, is assumed to be constant everywhere in the domain and it is also based on the measurements. The air density, $\rho$, is calculated from the equation of state $\rho = \frac{pR}{T}/m$, 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"), but with included 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 analysed time. The maximal timestep allowed by the Courant condition for convergence is $\Delta t = 10^{-6}$ s (Courant time step). Since $\Delta t$ is much larger than the time step needed to move the smallest particle to the neighbour size bin ($\Delta t_p = 2 \times 10^{-7}$ s), at every Courant time step we make 5 substeps 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 $\Delta t$ 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 Navier-Stokes system including equation for conservation of mass, momentum, energy and chemical species. In the present study we modify the model, describing the evolution of aerosol number density function: water vapour pressure over a droplet of radius $r$ is defined as (Seinfeld and Pandis, 2006; Sorjamaa and Laaksonen, 2007)

$$p_{w} = p_{0} \exp \left( \frac{A}{2r} - \frac{0.1d_{w}}{2(r-r_{0})} \right)$$

where $p_{0}$ is water vapor pressure over a flat surface at the same temperature, $A = 0.66/T$ (in $\mu m$), $r_{0} = 10$ 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 forces $f$ is used in a form

$$f(x,t) = Re\{N_{f_{k}}(t) \exp[i(k \cdot x + i\phi(t))]\},$$

where $x$ is the position vector. The wave vector $k(t)$ and the random phase $-\pi < \phi(t) \leq \pi$ change at every time step; $N = \int_{0}^{c_{v}}(|c_{v}|/\Delta t_{s})^{1/2}$ is the normalization factor, $c_{v}$ is the sound speed, $f_{k}$ is a non-dimensional forcing amplitude; $f_{k} = (k \times \mathbf{e})/\sqrt{k^{2} - (k \cdot \mathbf{e})^{2}}$, where $\mathbf{e}$ is an arbitrary unit vector that is real and not aligned with $k$ (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 (unequilibrium case), taking the environment temperature $T_{0} = 285.4$ K for equilibrium case and $T_{0} = 293$ K for equilibrium case. The buoyancy force is expressed as follows

$$B = g \left[\frac{T - T_{0}}{T_{0}} + \epsilon(Y_{H_{2}O} - q_{0}) - q_{c}\right],$$

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 (Andrejcuk et al., 2004; Babkovskaia et al., 2015). The summary of four considered cases is in Table 2.

3 Results

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

In Fig. 1 we present the air temperature averaged in $y$-direction. In Fig. 1 (a, b, c) we present the temperature distribution in the cases where the aerosol dynamics is included. We find that without aerosol (case 4) after 3 s the difference...
Fig. 3. Relative distribution of particles averaged over the domain at \( t = 0 \) s (black dotted curve) and \( t = 3 \) s (solid curve) for the cases of \( N_{\text{tot}} = 55.5 \) cm\(^{-3}\) (green solid curve), \( N_{\text{tot}} = 555 \) cm\(^{-3}\) (red solid curve), \( N_{\text{tot}} = 5550 \) cm\(^{-3}\) (black solid curve).

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

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

Table 3. Total number of particles (\( N_{\text{tot}} \)), number of activated particles (\( N_{\text{act}} \)), liquid water content (LWC), change of the temperature (\( \Delta T \)), maximal values of supersaturation (\( \Delta S_{\text{max}} \)), minimal values of supersaturation (\( \Delta S_{\text{min}} \)) at \( t = 3 \) s for three considered cases. 

| Case          | \( N_{\text{tot}} \), cm\(^{-3}\) | \( N_{\text{act}} \), cm\(^{-3}\) | LWC, g/cm\(^3\) | \( \Delta T \), K | \( \Delta S_{\text{max}} \), % | \( \Delta S_{\text{min}} \), % |
|---------------|-------------------------------|---------------------------------|-----------------|-------------------|----------------|----------------|
achieves maximum. It happens because of the different effect of aerosol dynamics in dependence of \( N_{\text{tot}} \). 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. 1, 2 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 grows 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 (\( S = 0 \%) \) in dependence on different total numbers of droplets to answer the question how \( N_{\text{tot}} \) affects the phase relaxation time. Analysing Fig. 2 we find that in a case of \( N_{\text{tot}} = 55 \text{ cm}^{-3} \) 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 (\( \tau_{\text{eddy}} = 2 \) s) (see discussion in section 2). Thus, strong supersaturation fluctuations can “survive” longer if the total number of droplets is smaller.

**Effect of total number of particles on activation**

In this study we consider that all particles with radius larger than \( r_{cr} = 1.75 \mu m \) are activated. This value is somewhat arbitrary but the results of our study were not sensitive on the choice of \( r_{cr} \), provided that \( r_{cr} \leq 1.75 \mu m \). In Fig. 3 we present the normalized particle distribution at \( t = 0 \) s and \( t = 3 \) s. We find that the smaller is 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. 2) one can see that in the case of the largest \( N_{\text{tot}} \) the supersaturation appears to be close to zero and particles stop to grow, while for the smaller \( N_{\text{tot}} \) the supersaturation is about 5% and particles continue growing. Therefore, the system is coming to an equilibrium (i.e. \( S = 0 \)) faster and aerosol particles cease to grow earlier for the larger \( N_{\text{tot}} \). Thus, our DNS results for a case without vertical displacement and resulting convective cooling confirm the Twomey effect and show that aerosol particles can grow and possibly achieve the size of the raindrop 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 condensational growth of drops, in order for rain to form (e.g. Rogers and Yau 1989,

![Fig. 6. Averaged vertical velocity as a function of time with aerosol for \( N_{\text{tot}} = 5500 \text{ cm}^{-3} \) (dotted curve); and without aerosol (solid curve). Unequilibrium and low intensive turbulence case is considered (see Table 3).](image1)

![Fig. 7. The dependence of the average kinetic energy on time with aerosol for \( N_{\text{tot}} = 5500 \text{ cm}^{-3} \) (dotted curve); and without aerosol (solid curve). The other parameters correspond to unequilibrium case with low intensive turbulence (see Table 2).](image2)
Fig. 8. The difference between turbulent kinetic energy averaged over time period $t$ in a case without aerosol ($\overline{T KE_{0}}$) and with aerosols ($\overline{T KE}$), where $\overline{T KE}$ = $\frac{1}{t} \int_{0}^{t} T KE(t') dt'$. 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 $N_{tot} = 5500$ cm$^{-3}$.
without them. We conclude that turbulence is damped because of presence of aerosols. We interpret these results as follows. The air temperature increases because of latent heating caused by condensation onto drops, and the temperatures inside and outside the domain is reduced. It results in decreased buoyancy force and deceleration in vertical direction. Thus, we conclude that aerosol affects the turbulence via buoyancy.

Also, we find that deceleration does not depend on intensity of turbulent fluctuations, i.e. deceleration in vertical direction is the same in low intensive turbulence and high intensive turbulence cases. Moreover, turbulent fluctuations are damped because of presence of aerosol particles in all four considered cases. However, the damping effect is stronger in unequilibrium with low intensive turbulence than in other three cases. The dependences of vertical velocity and turbulent kinetic energy (TKE) averaged over the domain as a function of time for unequilibrium 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 buoyancy force with time, which results in perturbation of vertical motion.

4 Summary

Turbulence, aerosol growth and microphysics of hydrometeors in clouds are intimately coupled. In the present study a new modelling approach was applied so as to quantify this linkage. 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. We study the interaction in the cloud area under transient, high supersaturation conditions, using direct numerical simulations. As the initial conditions we take the observational data obtained by the ACTOS platform. To analyse the effect of aerosol on the turbulence we choose the part of the cloud where the supersaturation is about 10% and condensation is the dominant process. As an initial distribution of particles we take the data of measurements at the sea level and analyse the drops activated by the aerosols in simulations.

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$^{-3}$) increases 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. Also, we find that the larger is $N_{eq}$ the smaller is the relative number of activated particles (averaged over the domain). Analysing the effect of aerosol dynamics on the turbulent kinetic energy and on vertical velocity we conclude that the presence of aerosol has an effect on vertical motion and in our case tends to reduce downward 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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