Impact of Giant Sea Salt Aerosol Particles on Precipitation in Marine Cumuli and Stratocumuli: Lagrangian Cloud Model Simulations

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ABSTRACT: The impact of giant sea salt aerosols released from breaking waves on rain formation in marine boundary layer clouds is studied using large-eddy simulations (LES). We perform simulations of marine cumuli and stratocumuli for various concentrations of cloud condensation nuclei (CCN) and giant CCN (GCCN). Cloud microphysics are modeled with a Lagrangian method that provides key improvements in comparison to previous LES of GCCN that used Eulerian bin microphysics. We find that GCCN significantly increase precipitation in stratocumuli. This effect is strongest for low and moderate CCN concentrations. GCCN are found to have a smaller impact on precipitation formation in cumuli. These conclusions are in agreement with field measurements. We develop a simple parameterization of the effect of GCCN on precipitation, accretion, and autoconversion rates in marine stratocumuli.

SIGNIFICANCE STATEMENT: Breaking sea waves release salt particles into the atmosphere. Cloud droplets formed on these salt particles can grow larger than droplets formed on other smaller particles. Therefore, sea salt particles can be important for rain formation over oceans. To investigate this effect, we performed idealized computer simulations of stratocumulus and cumulus clouds. Sea salt particles were modeled with an unprecedented precision thanks to the use of an emerging modeling method. In our simulations sea salt particles significantly enhance rain formation in stratocumuli, but not in cumuli. Our study has implications for climate models, because stratocumuli are important for Earth's energy budget and for rain enhancement experiments.

KEYWORDS: Cloud microphysics; Large eddy simulations; Aerosol-cloud interaction; Cloud parameterizations; Aerosol hygroscopicity

1. Introduction

It has long been suggested that warm-rain formation is affected by giant cloud condensation nuclei (GCCN) (Houghton 1938). There is no strict definition of GCCN, but most authors use this term for particles with dry radii greater than 1 μm . Cloud droplets formed on such large CCN grow by condensation of water vapor to radii exceeding 20 μm . Since collision efficiency sharply increases with droplet size, droplets formed on GCCN collide frequently with other droplets, including smaller droplets formed on smaller CCN. However, it is not clear how important this process is because concentrations of GCCN are typically two to three orders of magnitude lower than concentrations of smaller CCN. GCCN can come from various sources, but in this paper we focus on GCCN made of sea salt because large sea salt particles are ubiquitous over the oceans. They are produced by breaking waves and their concentration increases with the increase of the wind speed near the ocean surface (Woodcock 1953).

Because of the large solute mass in droplets formed on GCCN, their condensational growth differs from that of droplets with submicron CCN. The activation radius of droplets formed on GCCN can be hundreds of microns, and they can grow by condensation in downdrafts (Jensen and Nugent 2017). Therefore, the often made assumption that the solute effect can be neglected above the activation zone is not valid for droplets formed on GCCN.

Numerical models have been extensively used to study sensitivity of warm-rain production to GCCN. An important limitation of most of these studies is that they use simplified modeling frameworks. Most of them apply parcel models (Johnson 1982; Cooper et al. 1997; Segal et al. 2004; Jensen and Lee 2008; Lowenstein et al. 2010; Cooper et al. 2013; Jensen and Nugent 2017), some use 1D (Zhang et al. 2006), others 2D models (Yin et al. 2000; Blyth et al. 2013). These simplified models are useful for showing qualitatively that GCCN are important for rain formation, but quantitative assessments should be done using LES of a cloud field because only then a realistic representation of the interplay between cloud dynamics and microphysics is included. So far, only a handful of simulations of the effect of GCCN were done using LES (Feingold et al. 1999; Lu and Seinfeld 2005; Cheng et al. 2009; Kogan et al. 2012). However, these simulations, similarly to the parcel, 1D, and 2D simulations, used Eulerian bin microphysics schemes. Although bin schemes represent an established method of the detailed microphysics

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modeling, it has key drawbacks (Grabowski et al. 2019) that are particularly important for GCCN modeling. First, in Eulerian bin schemes, it is difficult (if not impossible) to model the effect of dissolved aerosol on condensational growth of cloud droplets. This effect is crucial for droplets formed on GCCN (Jensen and Nugent 2017). The solute effect can only be modeled in a 2D bin scheme; see Fig. 4 in Grabowski et al. (2019). Such a scheme requires very intensive computations, making it practically impossible to use in LES. Second, bin schemes are troubled by an artificial broadening of the droplet size spectra caused by numerical diffusion (Rémillard et al. 2017; Morrison et al. 2018; Witte et al. 2019; Lee et al. 2019; Grabowski 2020; Lee et al. 2021). This broadening can lead to an increase in the amount of precipitation produced solely by droplets formed on CCN, in which case GCCN have a smaller impact. Last, bin models often apply parameterization of droplet activation. These parameterizations have to be adjusted to properly represent possible effects of GCCN.

Most of the studies have shown that GCCN increase warm-rain production, but to various extent and under various GCCN concentrations. Feingold et al. (1999) have found that GCCN concentration of 10^{-3} cm^{-3} is enough to increase the amount of rain in stratocumuli by up to 100%. Jensen and Nugent (2017) predict an increase in the amount of rain in stratocumuli for a GCCN concentration of around 0.3 cm^{-3} . Other studies found little impact of GCCN on rain production in marine stratocumuli (Lu and Seinfeld 2005; Zhang et al. 2006). These authors did find that GCCN can enhance rain production, but only for high LWP and CCN concentrations exceeding 1000 cm^{-3} (Lu and Seinfeld 2005), or for high GCCN concentrations on the order of 60 cm^{-3} (Zhang et al. 2006). Moreover, Zhang et al. (2006) concluded that ambient GCCN concentrations, around 3 cm^{-3} , inhibit rain production. In cumulus clouds, parcel model results suggest that precipitation is sensitive to GCCN (Johnson 1982; Cooper et al. 1997; Segal et al. 2004; Lowenstein et al. 2010). Results of flow-resolving models are less conclusive. Cheng et al. (2009) report that as GCCN concentration is increased from 0.01 to 0.1 cm^{-3} , surface precipitation is increased by 10% in clean conditions (CCN concentration of 100 cm^{-3}) and by 100% in polluted conditions (CCN concentration of 1000 cm^{-3}). Kogan et al. (2012) found that in clean conditions (CCN concentration of around 100 cm^{-3}), GCCN at a concentration of 1.5 cm^{-3} increase the accumulated precipitation by around 30%. Finally, Blyth et al. (2013) did not find any significant effect of GCCN on rain in cumuli.

Many of the numerical studies agree that the effect of GCCN on precipitation development is modulated by the concentration of CCN, but there are discrepancies in the nature of this relationship. On one hand, in studies of Lu and Seinfeld (2005) and Cheng et al. (2009) the impact of GCCN increases with increasing CCN concentration, even over 1000 cm^{-3} . Feingold et al. (1999) and Jensen and Lee (2008) on the other hand show a maximum relative impact of GCCN for moderate CCN concentrations. The argument for that is that at low CCN concentrations, few cloud droplets are activated, they grow large through vapor condensation and

coalescence is efficient even without GCCN. For high CCN concentrations, cloud droplets formed on CCN are so small that they are not efficiently collected by large cloud droplets formed on GCCN. Therefore GCCN are expected to have the most impact for moderate CCN concentrations. In such a case, cloud droplets formed on CCN are large enough to be collected by larger droplets formed on GCCN, but not large enough to form significant precipitation on their own.

We summarize the modeling studies of GCCN done so far by citing Cooper et al. (2013): “The rain produced is sensitive to giant aerosols, but modification of the modeling framework is required to conduct a more robust test of their relative importance.” The goal of this work is to perform such robust tests with an improved modeling framework. We perform LES of marine clouds for various GCCN and CCN concentrations. The GCCN concentrations considered are in the range from around 0.2 to around 2 cm^{-3} . Such concentrations can be produced by breaking sea waves for wind speeds of up to 19 m s^{-1} (O’Dowd et al. 1997). We perform simulations for three values of CCN concentration in order to capture the modulation of impact of GCCN by CCN that was discussed earlier in this section. For each case, we average results over a small ensemble of simulations. We model cloud microphysics with a Lagrangian particle-based scheme. Because of that, we overcome the problems with Eulerian bin microphysics discussed above. Formation of new GCCN by sea waves is accounted for in the model by adding new GCCN throughout the simulation in the lower part of the domain.

Breaking sea waves release not only sea salt GCCN, but also smaller, submicron sea salt CCN. Recent measurements and parcel model simulations show that submicron sea salt CCN also can increase precipitation (Fossum et al. 2020). The reason is that, in some range of updraft speeds and CCN concentrations, less CCN are activated if both sulfate and sea salt CCN are present compared to conditions with sulfate CCN only. While this effect is not the main focus of our study, we perform some simulations with submicron sea salt CCN to check if their presence somehow modulates the impact GCCN have on precipitation.

2. Methods

a. Basic description of the model

The LES model used in this study is the University of Warsaw Lagrangian Cloud Model (UWLCM). UWLCM is an anelastic fluid flow model with Lagrangian particle-based cloud microphysics. Advection of Eulerian variables is modeled with the MPDATA algorithm (Smolarkiewicz 2006). The cloud microphysics model is an implementation of the superdroplet method (SDM) (Shima et al. 2009). Liquid water particles, including humidified aerosols, are represented in a Lagrangian fashion by computational point particles called superdroplets. Details of the numerical implementation of MPDATA and of SDM can be found in Jaruga et al. (2015) and in Arabas et al. (2015), respectively. In the presented simulations, the subgrid-scale (SGS) diffusion of Eulerian fields is done with the Smagorinsky–Lilly model (Lilly 1962). To

represent SGS transport of liquid water, each superdroplet is characterized by its own SGS velocity. Assuming that the SGS turbulence is homogeneous and isotropic, SGS velocity can be described with the Ornstein–Uhlenbeck process. In this simple approach, components of the SGS velocity are evolved according to Eq. (10) in Grabowski and Abade (2017). SGS fluctuations of supersaturation are not modeled, nor is the effect of SGS turbulence on collisional growth of droplets. Drop breakup is not included in the model. The change of droplet sizes due to condensation and evaporation is modeled with the Maxwell–Mason equation (Arabas et al. 2015). Water activity is parameterized with the κ parameter (Petters and Kreidenweis 2007). To make sure that the κ parameterization is valid for GCCN, we reproduced the parcel model simulations from Jensen and Nugent (2017), in which this parameterization was not used. A more detailed description of UWLCM can be found in Dziekan et al. (2019).

The key novelty of UWLCM is the use of Lagrangian microphysics. Lagrangian microphysics provide a level of detail and computational complexity similar to Eulerian bin microphysics, but offer several advantages (Grabowski et al. 2019). Advantages of Lagrangian microphysics that are crucial in this study are the lack of numerical diffusion and the ease of modeling additional droplet characteristics. Ease of adding more droplet characteristics means that it is computationally feasible to track CCN mass in each droplet. This is required to include, at all times, the solute effect in the equation for droplet condensational growth. Details of condensational growth of sea salt GCCN and the role played by the solute effect are discussed in detail in Jensen and Nugent (2017). Another important aspect of the microphysics scheme used in UWLCM is that all aerosol particles are followed, and the growth of unactivated CCN is modeled in the same way as of activated droplets. Because of that, no droplet activation parameterization is needed, and aerosol processing is simulated properly.

UWLCM was compared with other LES models in a stratocumulus simulation (Dziekan et al. 2019). The only major difference found was that UWLCM gives very little surface precipitation, less than most other models and less than expected from observations. We attribute this to the use of Lagrangian microphysics in UWLCM. The models with which UWLCM was compared use Eulerian microphysics, in which numerical diffusion can increase the amount of rain. In UWLCM, there is no artificial broadening of DSD. To make sure that it is the artificial broadening of DSD that is responsible for the differences in precipitation in Sc, Lagrangian and bin microphysics would need to be compared using the same model for dynamics. This could be a focus of a future study. Lower surface precipitation in UWLCM than observed during the field campaign suggests that some factors important for rain production, e.g., GCCN or the effects SGS turbulence has on condensation and collision–coalescence, were not included in the simulations presented in Dziekan et al. (2019). This leads us to believe that UWLCM is a proper tool for studying the impact of GCCN.

b. Description of the marine stratocumulus simulations

Marine stratocumulus simulations presented in this study follow the simulation setup described in Ackerman et al. (2009). This setup is based on observations of a drizzling nocturnal stratocumulus during the research flight 2 of the Second Dynamics and Chemistry of the Marine Stratocumulus field study (DYCOMS II) (Stevens et al. 2003). Initial temperature and moisture profile are shown in Fig. 1. Total simulation time is 6 h. The first hour is the simulation spinup with no collision–coalescence. The domain size is $6.4 \text{ km} \times 6.4 \text{ km} \times 1.5 \text{ km}$ with a uniform grid of $50 \text{ m} \times 50 \text{ m} \times 5 \text{ m}$. Numerical time steps were determined with convergence tests. The model time step is 1 s. Substepping with a 0.1-s time step is used for condensation and collision–coalescence.

c. Description of the marine cumulus simulations

The marine cumulus simulations apply the modeling setup based on observations from the Rain in Cumulus over the Ocean (RICO) field campaign (Raubert et al. 2007; VanZanten et al. 2011). This setup is widely used in LES of precipitating marine cumulus field [Sulak et al. (2020), Druke et al. (2020), Radtke et al. (2021), and Dixit et al. (2021) are some recent examples]. The domain is $12.8 \text{ km} \times 12.8 \text{ km} \times 4 \text{ km}$ with the grid spacing of $100 \text{ m} \times 100 \text{ m} \times 40 \text{ m}$. The simulations are run for 10 h. The model time step is 0.5 s. Convergence tests show that substepping for condensational or collisional growth is not necessary.

d. Aerosol size distribution

CCN and GCCN size distributions are plotted in Fig. 2. Simulations were done for seven different values of CCN concentration, listed in Table 1. In six cases all CCN are ammonium sulfate particles and there is one case in which CCN are a mixture of ammonium sulfate and submicron sea salt particles. The case with submicron sea salt CCN is meant to test how small sea salt particles released by breaking waves affect precipitation and potentially modulate the effect of GCCN. Name of this case contains the suffix *_salt_CCN*. Initial ammonium sulfate CCN radius is defined by a bimodal lognormal distribution. Parameters of these lognormal modes are given in Table 2. The cumulus and stratocumulus sulfate CCN size distributions are based on VanZanten et al. (2011) and Ackerman et al. (2009), respectively.

The size distribution of sea salt aerosols is based on measurements performed during the VAMOS Ocean–Cloud–Atmosphere–Land Study (VOCALS) campaign. These measurements are given in Jensen and Nugent (2017) in the form of a table with concentrations of aerosols in 38 size bins for dry radii from 0.8 to $9 \mu\text{m}$. We will denote the total concentration of particles in this size range by N_{GCCN} and refer to it as GCCN concentration. During the VOCALS campaign, $N_{\text{GCCN}} = 0.2817 \text{ cm}^{-3}$ was observed for a horizontal wind speed of 5.4 m s^{-1} . To represent sea salt aerosols in the model, a unimodal lognormal distribution was fitted to the VOCALS measurements. Parameters of this distribution are given in Table 2. The distribution is set to zero for dry

FIG. 1. Soundings used to initialize the simulations: liquid water potential temperature and total water mixing ratio for stratocumulus and potential temperature and water vapor mixing ratio for cumulus. Note that below the inversion in stratocumulus there is a layer of positive supersaturation.

radii exceeding $9 \mu\text{m}$, the maximum size for the [Jensen and Nugent \(2017\)](#) observed size distribution. Sea salt GCCN are assumed to be represented by the part of the distribution between 0.8 and $9 \mu\text{m}$. Sea salt CCN are assumed to be represented by the part of the distribution below $0.8 \mu\text{m}$.

Observations of the vertical gradient of sea salt loading in the marine atmosphere show only a very slight decrease with altitude in the mixed layer (excluding the immediate surface

zone), followed by a stronger decrease with altitude outside clouds in the trade wind cumulus layer. This is to be expected, as the sedimentation velocities of most hydrated GCCN are much lower than the average updraft/downdraft velocities in the mixed layer. [Figure 3](#) shows a typical profile of sea salt mixing ratio measured during RICO. To represent the observed vertical distribution of sea salts, simulations are started with sea salt GCCN and CCN uniformly distributed

FIG. 2. Aerosol size distributions used in (left) stratocumulus and (right) cumulus simulations. Orange lines are different concentrations of ammonium sulfate CCN. The solid, dashed, and dashed-dotted blue lines are different concentrations of sea salt GCCN. The dotted blue line in the left panel is the sea salt CCN distribution. Parameters of the aerosol lognormal modes are given in [Table 2](#).

TABLE 1. Initial concentrations of ammonium sulfate ($N_{\text{CCN}}^{\text{sul}}$) and sea salt CCN ($N_{\text{CCN}}^{\text{ss}}$) and the resulting mean cloud droplet concentration N_c . Cases are named by combining cloud type and the value of N_c . Cloud droplets are droplets with radii between 0.5 and 25 μm . N_c is averaged over cloudy cells from simulations without GCCN.

Case	$N_{\text{CCN}}^{\text{sul}}$ (cm^{-3})	$N_{\text{CCN}}^{\text{ss}}$ (cm^{-3})	N_c (cm^{-3})
Stratocumulus			
Sc38	95	0	38
Sc54_salt_CCN	190	19.6	54
Sc59	190 ^a	0	59
Sc115	475	0	115
Cumulus			
Cu38	52.5	0	38
Cu60	105 ^b	0	60
Cu88	210	0	88

^a As in Ackerman et al. (2009).

^b As in VanZanten et al. (2011).

below 460 m, which is below Sc and Cu cloud base. As the simulation progresses, sea salt particles are advected to higher parts of the domain.

Over the course of a long simulation aerosol concentrations can significantly decrease due to washout. To prevent this we model aerosol sources by relaxing aerosol concentration to the initial value. This approach is similar to the relaxation of horizontal wind components used in Grabowski et al. (1996) [see Eq. (8) there]. The idea is to introduce new aerosols whenever the average number of aerosols at a given level is lower than expected. Details of the algorithm are given in the appendix. Relaxation of sulfate and sea salt particles is done in the whole domain and below 460 m, respectively.

Efficacy of the relaxation approach can be seen in vertical profiles of CCN and GCCN concentrations (Fig. 4). Sulfate CCN concentration below the inversion is close to the initial value and does not change with time. GCCN concentration below 460 m is also close to the initial value and constant in time. Therefore the

initial GCCN concentration, which will be denoted by $N_{\text{GCCN}}^{\text{init}}$, is a good indicator of the actual GCCN concentration N_{GCCN} . In Cu, N_{GCCN} is constant in time also above the relaxation level, up to around 750 m. Above 750 m, N_{GCCN} steadily increases with time as GCCN from near the surface are advected by updrafts. In Sc, N_{GCCN} between 460 m and the inversion decreases with time. The reason is that for the first hour of the Sc simulation collision-coalescence is not modeled. During that time GCCN from below 460 m are advected up to the inversion. After the first hour, collision-coalescence is modeled and the droplets formed on GCCN start to grow larger resulting in faster sedimentation, which causes the decrease in concentration.

For each CCN concentration, we run a reference simulation without GCCN and 3 simulations in which varying concentrations of GCCN are added: $N_{\text{GCCN}}^{\text{init}} = 0.216 \text{ cm}^{-3}$, $N_{\text{GCCN}}^{\text{init}} = 1.08 \text{ cm}^{-3}$, and $N_{\text{GCCN}}^{\text{init}} = 2.16 \text{ cm}^{-3}$. The smallest GCCN concentration comes from the fit to the VOCALS data. Higher GCCN concentrations are meant to study the sensitivity of cloud precipitation to GCCN concentration. They can also be considered as cloud seeding experiments or as GCCN released from breaking waves at higher wind speeds. Judging from the parameterization of O'Dowd et al. (1997), the two higher $N_{\text{GCCN}}^{\text{init}}$ considered should be observed at wind speeds of around 12 and around 19 m s^{-1} near sea surface. However, Jung et al. (2015) observed 1.89 cm^{-3} particles with radii greater than 1 μm near sea surface already at a wind speed of 10 m s^{-1} .

The sea salt CCN concentration in the Sc54_salt_CCN test is associated with the highest GCCN concentrations considered herein, $N_{\text{GCCN}}^{\text{init}} = 2.16 \text{ cm}^{-3}$. That way we test a relatively strong effect of sea salt CCN. The resulting sea salt CCN concentration is around 20 cm^{-3} , compared with 25 cm^{-3} observed by Fossum et al. (2020) in an air mass with high sea salt CCN concentration.

In each model cell, the initial CCN spectrum is represented by 100 superdroplets. This number of superdroplets is sufficient to obtain correct averaged cloud characteristics, as shown by Arabas and Shima (2013) for cumuli and by Dziekan et al. (2019) for stratocumuli. The initial GCCN spectrum is represented by 40 superdroplets per cell. Sizes of

TABLE 2. Parameters of lognormal modes of the aerosol size distribution.

	Concentration	Mean radius (μm)	Geometric standard deviation	Hygroscopicity κ^a
Salt mode ^b	22.16 cm^{-3}	0.283	2.235	1.28
Stratocumulus				
Sulfate mode A ^c	$(125/190) \times N_{\text{CCN}}^{\text{sul}}$	0.011	1.2	0.61
Sulfate mode B ^c	$(65/190) \times N_{\text{CCN}}^{\text{sul}}$	0.06	1.7	0.61
Cumulus				
Sulfate mode A ^d	$(90/105) \times N_{\text{CCN}}^{\text{sul}}$	0.03	1.28	0.61
Sulfate mode B ^d	$(15/105) \times N_{\text{CCN}}^{\text{sul}}$	0.14	1.75	0.61

^a Values taken from Petters and Kreidenweis (2007).

^b Part of this mode greater (lower) than $r = 0.8 \mu\text{m}$ represents sea salt GCCN (CCN).

^c As in Ackerman et al. (2009).

^d As in VanZanten et al. (2011).

FIG. 3. Vertical profile of the sea salt mixing ratio (mass of sea salt per unit mass of air) from RICO measurements. Each point is one of microscope slides exposed from the NCAR/NSF C-130 research aircraft on 5 Jan 2005. For methodology, see [Jensen et al. \(2020\)](#). Slides were typically exposed for 5–60 s, depending on altitude. The average sample volume was 5 L s^{-1} .

superdroplets are initialized using an algorithm designed to correctly represent the size spectrum even for low number of superdroplets ([Dziekan and Pawlowska 2017](#)). The GCCN superdroplets have hygroscopicity parameter $\kappa = 1.28$. In the case of coalescence of superdroplets, particulate matter volumes are added and a new value of κ is calculated as a weighted arithmetic mean of κ of the coalescing superdroplets, in which weights are the volumes of the particulate matter ([Petters and Kreidenweis 2007](#)).

In addition to the main set of simulations, we also ran simulations with GCCN initially in the whole domain and without aerosol sources. To make it easy to distinguish these modeling cases from the main set of simulations, their names contain the suffix *_wash*, e.g., *Sc30_wash*. The results, shown in [section 2](#) of the online supplement, are consistent with results of the main set of simulations. This indicates that our conclusions are robust with regard to the assumptions about the vertical distribution of GCCN and that precipitation is mostly affected by GCCN that enter the cloud with updrafts going through cloud base.

e. Definitions

Here, we give definitions of characteristics discussed in [section 3](#). Cloud droplets are droplets with radii between 0.5 and $25 \mu\text{m}$. Rain drops are droplets with $r > 25 \mu\text{m}$. Autoconversion rate is the mass of rain droplets formed by collisions between two cloud droplets per unit time and unit volume. Accretion rate is the increase of mass of rain droplets caused by collisions between a cloud droplet and a rain drop per unit time and unit volume. Accretion and autoconversion rates are averaged over cloudy cells. In Sc, cloudy cells are those with

cloud droplet concentration greater than 20 cm^{-3} , as in [Ackerman et al. \(2009\)](#). In Cu, cloudy cells are those with cloud water mixing ratio greater than 0.01 g kg^{-1} , as in [VanZanten et al. \(2011\)](#). Surface and cloud-base precipitation rates are averaged over all columns, cloudy or not. Cloud cover is the fraction of columns with at least one cloudy cell. Cloud-base precipitation is calculated at the lowest altitude at which the liquid water mixing ratio averaged over a horizontal plane is greater than 0.1 g kg^{-1} in Sc and 0.005 g kg^{-1} in Cu.

Time-averaged values of surface precipitation rate, cloud-base precipitation rate, autoconversion rate, and accretion rate are denoted by P_{surf} , P_{clb} , R_{acnv} , and R_{accr} , respectively. The averaging period is 2–6 h in Sc and 3–10 h in Cu. These periods were chosen based on the criterion that the cloud droplet concentration is quasi stationary. No surface precipitation develops before these averaging periods. Precipitation rates divided by cloud fraction are denoted by C_{surf} and C_{clb} , which are averaged over the same period.

3. Results

Presented results are based on small ensembles of simulation runs. Ensemble sizes are included in the list of simulations, which is provided in [section 3](#) of the supplement. To help readers visualize the modeled cloud fields, example cross sections are plotted in [Figs. 5 and 6](#). Overall, bulk cloud properties (e.g., LWP, cloud cover, cloud depth, vertical velocity variance, and cloud droplet concentration) agree with results presented in [Ackerman et al. \(2009\)](#) for stratocumulus and [VanZanten et al. \(2011\)](#) for cumulus. Time series of these properties are shown in [section 1](#) of the supplement.

One cloud property that differs from some other models and from measurements is surface precipitation in stratocumulus. For example, in *Sc59* simulations without GCCN UWLCM predicts no surface precipitation, while the expected surface precipitation based on measurements is at least 0.24 mm day^{-1} ([Ackerman et al. 2009](#)). As will be shown, including realistic concentrations of GCCN in Sc simulations leads to a significantly better agreement in the amount of rain between UWLCM and field observations. Some of the other LES models that use bin microphysics give rain amounts close to observations ([Ackerman et al. 2009](#)), but this may be for the wrong reasons as bin microphysics give too much drizzle due to numerical diffusion. Regarding Cu, it is difficult to decisively compare modeled rain amount with observations ([VanZanten et al. 2011](#)) because the initial sounding is an idealization of the mean conditions during the weakly precipitating period during RICO field project. To make our analysis of the effect of GCCN independent of uncertainties regarding the amount of rain produced even without GCCN, we present results for different CCN concentrations that modulate the amount of rain.

Because we are concerned here with the impact of GCCN on precipitation, we focus on the autoconversion and accretion rates and on the rain rates at the surface and at the cloud base. To obtain a systematic understanding of the GCCN impact, we plot these characteristics as a function of the

FIG. 4. Vertical profiles of sea salt GCCN (droplets with dry radius exceeding $0.8 \mu\text{m}$) and sulfate CCN concentrations plotted every 2 h. Results of the *Sc59* and *Cu60* simulations for $N_{\text{GCCN}}^{\text{init}} = 2.16 \text{ cm}^{-3}$. The dashed horizontal line indicates the level below which relaxation of the GCCN concentration is done.

GCCN concentration for different CCN concentrations. In most plots, precipitation rates are divided by cloud cover. This is done to determine the increase in precipitation in clouds and not in the entire domain. It also allows a direct comparison between Sc, in which cloud cover is between 80% and 100%, and Cu, in which cloud cover is between 20% and 30%. Cloud cover can be decreased to some extent by GCCN. The maximum relative decrease is $\sim 10\%$ (Fig. 1h in the supplement).

a. Stratocumulus results

In *Sc59* and *Sc115* precipitation, accretion and autoconversion rates do not vary much in time in the 2–6-h period (Figs. 2 and 3 in the supplement). In *Sc38* these parameters slowly increase with

time, but the rate at which they increase is the same for all values of $N_{\text{GCCN}}^{\text{init}}$ (Fig. 1 in the supplement). Therefore in all cases precipitation changes induced by GCCN are constant in time.

The average precipitation rates at the surface and at the cloud base as well as the average accretion and autoconversion rates are plotted in Fig. 7. A clear increase of all these characteristics is seen as GCCN concentration goes up. Even a rather low concentration of GCCN, $N_{\text{GCCN}}^{\text{init}} = 0.216 \text{ cm}^{-3}$, can increase C_{surf} by up to 0.08 mm day^{-1} (Fig. 7a). The magnitude of this increase depends on the concentration of CCN: it is the largest for the smallest CCN concentration (*Sc38*) and the smallest for the largest CCN concentration (*Sc115*). However, the relative impact of GCCN is arguably the largest for moderate CCN concentration (*Sc59*). In that case there is

FIG. 5. Cross sections at 6 h from the *Sc59* simulation with $N_{\text{GCCN}}^{\text{init}} = 2.16 \text{ cm}^{-3}$. Spatial interpolation was done to improve visual clarity.

very little surface precipitation without GCCN, but a non-negligible amount even for low GCCN concentration. For *Sc38* significant amount of rain is produced even without GCCN. In *Sc59* and *Sc38* the slope of the precipitation-GCCN relationship decreases with increasing number of GCCN. Namely, there is a bigger difference between $N_{\text{GCCN}}^{\text{init}} = 0 \text{ cm}^{-3}$ and $N_{\text{GCCN}}^{\text{init}} = 1 \text{ cm}^{-3}$ than between $N_{\text{GCCN}}^{\text{init}} = 1 \text{ cm}^{-3}$ and $N_{\text{GCCN}}^{\text{init}} = 2 \text{ cm}^{-3}$. In the *Sc115* case precipitation increases approximately linearly with $N_{\text{GCCN}}^{\text{init}}$ in the considered range of $N_{\text{GCCN}}^{\text{init}}$.

The *Sc54_salt_CCN* simulation has the same concentration of sulfate aerosols as *Sc59*, but with additional 19.6 cm^{-3}

submicron sea salt aerosols. Including these salt CCN results in a decrease of cloud droplet concentration by approximately 5 cm^{-3} , from $N_c \approx 59 \text{ cm}^{-3}$ to $N_c \approx 54 \text{ cm}^{-3}$, as shown in Fig. 8. Fossum et al. (2020) measured two air masses with different sea salt CCN concentrations and concluded that sea salt CCN decrease concentration of cloud droplets, which is in qualitative agreement with our modeling results. Comparing *Sc59* with *Sc54_salt_CCN* we see that the decrease in cloud droplet concentration does not affect precipitation much. However, in some cases such decrease of N_c can have visible impact on precipitation as seen in the *Sc40_salt_CCN_wash* simulations presented in section 2 of the supplement. Therefore not only

FIG. 6. Cross sections at 10 h from the *Cu60* simulation with $N_{\text{GCCN}}^{\text{init}} = 2.16 \text{ cm}^{-3}$.

FIG. 7. Mean precipitation rates at the (a) surface and at (b) the cloud-base altitude, both divided by cloud cover, and (c) accretion and (d) autoconversion rates vs GCCN concentration from the marine stratocumulus simulations. Definitions of these characteristics are given in [section 2e](#). Points show the mean from an ensemble of simulations. Error bars show an estimator of the standard error of the mean $SME = \sigma/\sqrt{n}$, where σ is the ensemble standard deviation and n is the ensemble size.

sea salt GCCN, but also sea salt CCN have the potential to increase the amount of precipitation. Our results suggest that the increase induced by sea salt CCN is smaller than the one caused by sea salt GCCN. Regarding our analysis of the impact of GCCN, sea salt CCN have little importance because a similar decrease in N_c as the one caused by salt CCN can be achieved by decreasing concentration of ammonium sulfate CCN.

It is instructive to compare our modeling results with the stratocumulus cloud seeding experiment reported by [Jung et al. \(2015\)](#). In that experiment, 0.5–5- μm radius salt particles

were released within cloud, near cloud top. The concentration of released particles was estimated to be from 10^{-2} to 10^{-4} cm^{-3} . Before cloud seeding, the mean cloud droplet concentration was 143 cm^{-3} and the mean effective radius measured near cloud top was around $11 \mu\text{m}$. These preseeding conditions resemble our *Sc115* case, in which the respective values are 115 cm^{-3} and $11.6 \mu\text{m}$. In the experiment, mean precipitation rate at cloud base before seeding was 0.04 mm h^{-1} . In our simulations without GCCN, C_{clb} is an order of magnitude smaller ([Fig. 7b](#)). However, [Jung et al. \(2015\)](#) observed that near sea surface there were 1.89 cm^{-3}

FIG. 8. Time series of cloud droplet concentration in stratocumuli for $N_{\text{GCCN}}^{\text{init}} = 0 \text{ cm}^{-3}$. Cloud droplet concentrations are averaged over cloudy cells. Two modeling cases are compared that differ only in the amount of submicron sea salt CCN. Only the averaging period 2–6 h is shown, during which cloud droplet concentration is quasi stable.

ambient CCN with $r > 1 \mu\text{m}$. If such $N_{\text{GCCN}}^{\text{init}}$ is assumed in our model, C_{clb} is around 0.038 mm h^{-1} , much closer to the observed value. This value was obtained by a linear interpolation of C_{clb} between results for $N_{\text{GCCN}}^{\text{init}} \approx 1.08 \text{ cm}^{-3}$ and $N_{\text{GCCN}}^{\text{init}} \approx 2.16 \text{ cm}^{-3}$, plotted in Fig. 7b. Jung et al. (2015) report that for 1.89 cm^{-3} ambient GCCN near sea surface, seeding 10^{-2} cm^{-3} GCCN near cloud top resulted in a four-fold increase of cloud-base precipitation in the seeded region. This cannot be represented in simulations by increasing GCCN concentration near the sea surface in the entire domain from $N_{\text{GCCN}}^{\text{init}} = 1.89 \text{ cm}^{-3}$ to $N_{\text{GCCN}}^{\text{init}} = 1.9 \text{ cm}^{-3}$. Such an increase has a minuscule effect on the domain-averaged precipitation. To properly model the effect of seeding, GCCN would have to be added locally near cloud top in a pattern resembling flight path of the seeding aircraft.

Another comparison with observations can be done using the data from the DYCOMS II campaign. The expected domain-averaged precipitation in the DYCOMS II RF02 setup with the CCN concentration as in the Sc59 case is between 0.24 and 0.46 mm day^{-1} at the surface and between 1.16 and 1.43 mm day^{-1} at cloud base (Ackerman et al. 2009). Our simulations without GCCN give $P_{\text{surf}} \approx 0.004 \text{ mm day}^{-1}$ and $P_{\text{clb}} \approx 0.15 \text{ mm day}^{-1}$, much less than expected. The horizontal wind velocity near the surface is around 9.5 m s^{-1} . We consider two potential values of $N_{\text{GCCN}}^{\text{init}}$: 1.89 cm^{-3} , a value observed by Jung et al. (2015) for horizontal wind velocity of around 10 m s^{-1} and 0.82 cm^{-3} , a value calculated using the O'Dowd et al. (1997) parameterization. Then, P_{surf} (P_{clb}) in our model is around 0.22 mm day^{-1} (1.5 mm day^{-1}) and 0.13 mm day^{-1} (1.1 mm day^{-1}), respectively. These values

were estimated by linear interpolation of P_{surf} and P_{clb} from simulations. Note that P_{surf} and P_{clb} are not plotted, but are around 85% of C_{surf} and C_{clb} that are plotted in Figs. 7a and 7b. We conclude that the discrepancy in the amount of precipitation between our LES results and observations are significantly reduced by including the effect of GCCN at realistic concentrations.

b. Cumulus results

Depth of the modeled cumulus clouds increases with time, up to around 1.3 km at the end of the simulation (Figs. 4–6 in the supplement). As clouds get deeper with time LWP increases and, in particular for low cloud droplet concentrations, RWP increases as well. Figure 9 depicts how GCCN affect precipitation, accretion and autoconversion rates averaged over the 3–10-h period. Accretion rate is increased by GCCN. The maximum increase for $N_{\text{GCCN}}^{\text{init}} = 0.216 \text{ cm}^{-3}$ is around $0.5 \text{ g m}^{-3} \text{ day}^{-1}$ observed for moderate CCN concentration (Fig. 9d, Cu60). The effect of GCCN on autoconversion varies depending on CCN concentration (Fig. 9c). GCCN increase autoconversion for high CCN concentration (Cu88), but only by up to around $0.05 \text{ g m}^{-3} \text{ day}^{-1}$ for $N_{\text{GCCN}}^{\text{init}} = 2.16 \text{ cm}^{-3}$. For moderate CCN concentration (Cu60), GCCN do not affect autoconversion and for low CCN concentration (Cu38) GCCN decrease autoconversion, by up to around $0.07 \text{ g m}^{-3} \text{ day}^{-1}$ for $N_{\text{GCCN}}^{\text{init}} = 2.16 \text{ cm}^{-3}$. Cloud-base precipitation increases with GCCN concentration, but the effect is rather weak (Fig. 9b). For $N_{\text{GCCN}}^{\text{init}} = 2.16 \text{ cm}^{-3}$ the maximum increase of C_{clb} is around 0.3 mm day^{-1} (Cu38). However, more than 50% of the GCCN-induced additional cloud-base precipitation reaches the surface. For example, the 0.3 mm day^{-1} increase of C_{clb} gives an increase of C_{surf} of around 0.22 mm day^{-1} (Fig. 9a, Cu38). The Cu35_wash case shows that GCCN do not affect precipitation at all if cloud droplet concentration is low (Fig. B2a). The value of C_{surf} in that case is around 1.2 mm day^{-1} regardless of GCCN concentration.

Measurements done during the RICO campaign led to the conclusion that GCCN have little impact on precipitation in cumuli (Reiche and Lasher-Trapp 2010; Minor et al. 2011). In our simulations the impact of GCCN on autoconversion, accretion and cloud-base precipitation in cumuli is also small, significantly smaller than in stratocumuli. However, surface precipitation in cloudy columns is increased by roughly half of the amount it is increased in stratocumuli.

c. Differences between Sc and Cu results

There are important differences in the way GCCN affect precipitation in Sc and Cu. In this section we will discuss these differences based on simulations with $N_{\text{GCCN}}^{\text{init}} = 2.16 \text{ cm}^{-3}$, because the impact of GCCN is most clearly seen in this high GCCN concentration case.

In Sc, R_{acnv} is increased by GCCN regardless of CCN concentration (Fig. 7c). The maximum increase, around $0.34 \text{ g m}^{-3} \text{ day}^{-1}$, is seen for low CCN concentration (Sc38). In Cu changes in R_{acnv} are smaller and their sign depends on CCN concentration (Fig. 9c). Their values vary from $-0.07 \text{ g m}^{-3} \text{ day}^{-1}$ for low CCN concentration (Cu38) to

FIG. 9. As in Fig. 7, but for the marine cumulus simulations.

$0.05 \text{ g m}^{-3} \text{ day}^{-1}$ for high CCN concentration (*Cu88*). Differences in autoconversion are also visible in time series of rain drop concentration [panel (f) in Figs. 1–3 and panel (g) in Figs. 4–6 in the supplement]. Rain drop concentration is increased in Sc by a value between 0.1 cm^{-3} (*Sc115*) and 0.15 cm^{-3} (*Sc38*). A much smaller increase of rain drop concentration is seen in Cu: 0.015 cm^{-3} in *Cu85* and 0.02 cm^{-3} in *Cu60*. For low CCN concentration, *Cu38*, GCCN do not increase rain drop concentration.

The value of R_{accr} is increased both in Cu (Fig. 9d) and in Sc (Fig. 7d), but by a smaller amount in Cu. In Sc that increase is highest ($7 \text{ g m}^{-3} \text{ day}^{-1}$) for low CCN concentration (*Sc38*) and lowest ($2.2 \text{ g m}^{-3} \text{ day}^{-1}$) for high CCN concentration (*Sc115*). In Cu the largest increase ($1.5 \text{ g m}^{-3} \text{ day}^{-1}$) is seen for moderate CCN concentration (*Cu60*).

GCCN cause a much bigger increase of cloud-base precipitation rate in Sc (Fig. 7b) than in Cu (Fig. 9b). In Sc, C_{clb} is increased by around 2 mm day^{-1} for *Sc38*, around

1.5 mm day^{-1} for *Sc59* and around 1 mm day^{-1} for *Sc115*. In Cu the maximum increase is only around 0.3 mm day^{-1} (*Cu38*). However, the drop-off in precipitation flux between cloud base and the surface is much smaller in Cu than in Sc. This makes the GCCN-induced differences in surface precipitation quite similar in Sc (Fig. 7a) and in Cu (Fig. 9a), despite the differences in the way cloud-base precipitation is affected. The maximum increase of C_{surf} is around 0.36 mm day^{-1} in Sc (*Sc38*) and around 0.22 mm day^{-1} in Cu (*Cu38*).

4. Parameterization of GCCN in marine stratocumuli

Looking for a generic description of the impact of wave released sea salt GCCN on marine stratocumuli, in Fig. 10 we plot Sc results as a function of the ratio of GCCN to CCN concentrations. We see that surface and cloud-base precipitation and accretion and autoconversion rates all follow a common pattern for different CCN concentrations. The only

FIG. 10. Mean precipitation rates at (a) the surface and at (b) the cloud-base altitude, and (c) accretion and (d) autoconversion rates from the marine stratocumulus simulations. Horizontal axis is the ratio of N_{GCCN}^{init} to $N_{CCN} = N_{CCN}^{sul} + N_{CCN}^{ss}$. P_{surf}^0 , P_{clb}^0 , R_{acnv}^0 , and R_{accr}^0 are values for $N_{GCCN}^{init} = 0 \text{ cm}^{-3}$. Lines show fitted power functions. The *Sc115* case was excluded from the fit to $P_{surf} - P_{surf}^0$.

exception is the surface precipitation in the high CCN concentration case, *Sc115*, which is consistently lower than in the other cases.

A simple power law can describe the impact of GCCN in stratocumuli:

$$X = X_0 + aG^b, \quad (1)$$

where X is either P_{surf} , P_{clb} , R_{acnv} , or R_{accr} ; X_0 is the value of X in the absence of GCCN; a and b are parameters fitted to the LES results; and $G = N_{GCCN}^{init}/N_{CCN}$ is the ratio of concentration of GCCN (aerosols with $r > 0.8 \mu\text{m}$) near

sea surface to CCN concentration. Values of parameters a and b are given in Table 3 and values of X_0 are given in Table 4.

This relationship could be used to parameterize the impact of GCCN on stratocumuli in numerical models. Concentration of GCCN can be calculated based on the wind speed near sea surface (O'Dowd et al. 1997). Such parameterization should be used with caution for high CCN concentrations, because the fitted function overestimates surface precipitation in *Sc115*, and for G exceeding 0.023, the maximum value modeled by us.

TABLE 3. Parameters of Eq. (1) fitted to the data in Fig. 10.

Characteristic	a	b
P_{surf}	2.2 mm day ⁻¹	0.51
P_{clb}	5.5 mm day ⁻¹	0.34
R_{acnv}	3.7 g m ⁻³ day ⁻¹	0.62
R_{accr}	63 g m ⁻³ day ⁻¹	0.57

We note that the cumulus results scaled in the same manner cannot be described with a single function. The surface and cloud-base precipitation rates and the accretion rate increase with G , but at different rates for different CCN concentrations. Depending on the CCN concentration, the autoconversion rate either decreases, remains constant, or increases with G .

5. Conclusions

We conducted LES that are a major step forward in the way the giant sea salt aerosols are treated in numerical models of clouds. Improvements over the previous LES studies include modeling of solute term in condensational growth of cloud droplets, no numerical diffusion of droplet sizes, different hygroscopicities of sea salt and sulfate aerosols, no parameterization of activation, and modeling of aerosol processing. Aerosol sources were modeled by relaxing CCN distribution to the initial one. Sea salt aerosols were added only in the lower part of the domain to mimic their release from the surface. Conclusions from the previous numerical studies of this subject range from no impact of GCCN to significant impact at very low GCCN concentrations, which indicates that further improvement of modeling methodology was needed. We believe that our model provides such an improvement. This belief is based on the fact that the model compares well with observations of sea salt aerosols in the following way. First, the amount of surface precipitation in stratocumulus simulations is in quantitative agreement with observations done by Jung et al. (2015), but only if GCCN are modeled. Second, in cumulus simulations the impact of GCCN on precipitation formation is smaller, which is in qualitative agreement with observations of Reiche and Lasher-Trapp (2010) and Minor et al. (2011). Third, our model also reproduced the recently observed effect that submicron sea salt CCN can decrease the number of cloud droplets (Fossum et al. 2020). Our study not only shows how sea salt GCCN affect precipitation in marine clouds, but also demonstrates that Lagrangian particle-based microphysics are well suited to model salt aerosols, which is important for cloud seeding experiments.

TABLE 4. Values of X_0 in Eq. (1).

Characteristic	$Sc38$	$Sc54_salt_CCN$	$Sc59$	$Sc115$
P_{surf} (mm day ⁻¹)	0.13	0.013	0.0037	7.7×10^{-6}
P_{clb} (mm day ⁻¹)	0.7	0.19	0.15	0.074
R_{acnv} (g m ⁻³ day ⁻¹)	0.26	0.041	0.019	1.7×10^{-4}
R_{accr} (g m ⁻³ day ⁻¹)	1.86	0.24	0.084	1.7×10^{-4}

In our simulations, wave-released sea salt GCCN were important for precipitation formation in marine stratocumuli. GCCN released at wind speeds around 5 m s⁻¹ can increase cloud-base precipitation in cloudy columns by up to 0.5 mm day⁻¹, surface precipitation in cloudy columns by up to 0.08 mm day⁻¹, autoconversion rate by up to 0.08 g m⁻³ day⁻¹ and accretion rate by up to 1.8 g m⁻³ day⁻¹. At higher wind speeds the effect is stronger. Changes caused by GCCN are largest for low cloud droplet concentrations and become smaller as cloud droplet concentration increases. However, even for 115 cm⁻³ cloud droplets the impact of GCCN is nonnegligible.

Precipitation formation in cumuli is less sensitive to GCCN. With increasing GCCN concentration, autoconversion rate increases for high N_c , is constant for moderate N_c and decreases for low N_c . Absolute value of the maximum change in autoconversion for a 5 m s⁻¹ wind is around 0.02 g m⁻³ day⁻¹. Accretion and precipitation rates increase with increasing GCCN concentration. For a 5 m s⁻¹ wind, accretion is increased by up to 0.5 g m⁻³ day⁻¹, cloud-base precipitation in cloudy columns by up to 0.08 mm day⁻¹ and surface precipitation in cloudy columns by up to 0.05 mm day⁻¹. GCCN have largest impact for moderate (60 cm⁻³) cloud droplet concentrations.

We found that the impact of GCCN on autoconversion, accretion, and precipitation in stratocumuli can be described with the same power law for different CCN concentrations. This power law can be used to include a parameterization of GCCN in large-scale models. This parameterization requires only two input parameters: concentrations of CCN and of GCCN. The latter can be calculated based on the horizontal wind speed near sea surface (O'Dowd et al. 1997).

There are strong reasons to believe that our results are representative of clouds formed in coupled PBL. The impact of GCCN is not expected to strongly depend on the cloud-base height, because GCCN are relatively uniformly distributed with altitude in most of the mixed layer. Differences in horizontal wind speeds were accounted for in our simulations by considering different GCCN concentrations. It is reasonable to expect that much less GCCN reach cloud base in a decoupled PBL, making GCCN less important for precipitation formation. However, additional numerical and observational studies would be needed to test this hypothesis.

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Data availability statement. Source code of UWLCM and of its dependencies libmpdata++ and libcloudph++ is available at <https://github.com/igfuw>. In the study, the following code versions were used: UWLCM v1.4 beta (Dziekan and Waruszewski 2021), libmpdata++ v1.3.0 (Jaruga et al. 2021), and libcloudph++ v2.1.2 beta (Arabas et al. 2021), which are available in Zenodo.

Data are available at Dziekan et al. (2021).

APPENDIX

CCN Relaxation Procedure for Lagrangian Particle-Based Microphysics

Aerosol processing is explicitly modeled in particle-based Lagrangian microphysics in which aerosols are represented with superdroplets. This may lead to significant changes over time of the aerosol distribution in LES. Often this is not desired and to prevent this, sources of aerosols need to be included in the model.

One approach is to parameterize activation of superdroplets (Grabowski et al. 2018). In it, a constant aerosols distribution is assumed and superdroplets represent cloud and rain drops only. Disadvantages of this approach are that activation is not modeled explicitly and that aerosol processing cannot be modeled.

For this work we developed a different approach, in which sources of aerosols are modeled by adding SD that represent relaxation of the aerosol distribution to the initial state. Similarly to the relaxation of horizontal wind components in Grabowski et al. (1996), the goal is to relax the horizontal average at each height and not the value in each individual model cell. Relaxation works only in one direction, i.e., new SD are added when there are too few aerosols, but we do not remove SD when the aerosol concentration exceeds the initial condition.

Input parameters of the relaxation procedure are time scale τ (10 min, on the order of the eddy turnover time), number of size bins N_{bin} (100), number of SD created per bin $N_{\text{bin}}^{\text{SD}}$ (400), and how often the procedure is applied t_{rel} (2 min). In parentheses are the values used in this study. In addition, user needs to define any number of size distributions toward which the aerosol concentration should be relaxed. We will refer to those as relaxation distributions. Each of these distributions is accompanied by the hygroscopicity parameter κ with which new aerosols are initialized, the range of κ of aerosols that are assumed to be a part of this distribution and by the altitude range in which relaxation toward this distribution should be done.

At initialization, range of radii of each relaxation distribution is determined by looking for the minimum and maximum radii, r_{min} and r_{max} , for which the expected number of droplets in a bin is greater than one (Dziekan and Pawlowska 2017). Next, the number of bins used for each distribution is calculated as $N_{\text{bin}} \times [\ln(r_{\text{max}}) - \ln(r_{\text{min}})] / \sum [\ln(r_{\text{max}}) - \ln(r_{\text{min}})]$, where summation is done over all relaxation distributions and the result is rounded to the nearest integer.

During the simulation the following algorithm is run every t_{rel} for each size bin of each relaxation distribution and at each height level within the range of heights of this distribution:

- 1) Calculate N_{tot} , the total number of droplets with dry sizes within the bin and with κ within the range of this distribution.
- 2) Calculate how many droplets should be added: $N_{\text{add}} = \max[0, (N_{\text{tot}} - N_{\text{exp}})] \times \min(1, t_{\text{rel}}/\tau)$, where N_{exp} is the expected number of droplets calculated from the relaxation distribution.
- 3) If $N_{\text{add}} > 0$, create $N_{\text{bin}}^{\text{SD}}$ superdroplets with κ equal to the initial κ for this relaxation distribution, dry radii randomly selected within the size bin, positions randomly selected at the height level and multiplicities equal to $N_{\text{add}}/N_{\text{bin}}^{\text{SD}}$ rounded to the nearest integer.
- 4) The wet radius of each created superdroplet is initialized to be in balance for the local relative humidity. If the local relative humidity exceeds 95%, the wet radius is initialized to be in balance with the 95% relative humidity.

The algorithm is run every t_{rel} and not every time step, because it is computationally expensive. Also in simulations with little washout, running the algorithm every time step would lead to creation of multiple SD with small multiplicities.

In the study we used two relaxation distributions, one for sulfate aerosols and one for sea salt aerosols. Parameters of these size distributions are given in Table 2. For the sulfate relaxation distribution the initial κ is 0.61, the range of κ is from 0.61 to 0.945 and the range of altitudes is the whole domain. For the sea salt relaxation distribution the initial κ is 1.28, the range of κ is from 0.945 to 1.28, and the range of altitudes is from the surface to 460 m.

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