We study the dispersion of sea salt particles from artificially injected sea spray at a cloud-resolving scale. Understanding of how different aerosol processes affect particle dispersion is crucial when designing emission sources for marine cloud brightening. Compared with previous studies, we include for the first time an explicit treatment of aerosol water, which takes into account condensation, evaporation and their effect on ambient temperature. This enables us to capture the negative buoyancy caused by water evaporation from aerosols. Additionally, we use a higher model resolution to capture aerosol loss through coagulation near the source point. We find that, with a seawater flux of $15 \text{ kg s}^{-1}$, the cooling due to evaporation can be as much as $1.4 \text{ K}$, causing a delay in particle dispersion of $10–20 \text{ min}$. This delay enhances particle scavenging by a factor of 1.14 compared with simulations without aerosol water. We further show that both cooling and particle dispersion depend on the model resolution, with a maximum particle scavenging efficiency of 20% within 5 h after emission at maximum resolution of 50 m. Based on these results, we suggest further regional high-resolution studies which model several injection periods over several weeks.
requires cutting down the emissions; however, the international political and social communities have so far been slow in adopting significant mitigation policies [2,3]. As an alternative, researchers have suggested several geoengineering methods [4] which deliberately modify the atmospheric radiative budget in order to reduce or counteract the climate effects of GHG emissions. Methods proposed aim to either reduce the incoming solar radiation or increase the outgoing thermal radiation. A decrease in incoming solar radiation can be achieved by increasing the Earth’s albedo using, for example, aerosol technologies that mimic either volcanic eruptions or natural sea spray production. Marine cloud brightening (MCB), i.e. artificially increasing sea salt particle concentration in marine environments in order to enhance marine cloud albedo through aerosol indirect effects, is considered to be one of the potential ways to counteract the warming effect of anthropogenic GHG emissions. MCB’s advantage over other geoengineering methods is that its effects are short-lived, which means that it can be easily and quickly terminated.

Low-level stratocumulus clouds cover a large area of the ocean surface and reflect solar radiation back into space, thus reducing the amount of solar radiation reaching the Earth’s surface. Latham [5] proposed that the albedo of marine boundary layer clouds could be increased enough to counteract warming from up to quadrupled CO₂ concentrations by injecting a high number concentration of sea salt particles into the air, which readily activate to cloud droplets. According to Latham et al. [6], a 0.06 increase in global albedo produces a \(-3.7\, \text{Wm}^{-2}\) radiative effect, which would be enough to counteract the positive forcing produced by doubled GHG concentration since the pre-industrial era. Salter et al. [7] proposed deploying a fleet of unmanned vessels that produce a flux of sea spray into the air at an emission rate of \(1.45 \times 10^6\, \text{m}^{-2}\, \text{s}^{-1}\) in order to achieve a 0.09 increase in cloud albedo through the Twomey effect [8]. They concluded that a cloud droplet number concentration (CDNC) of \(373\, \text{cm}^{-3}\) globally is needed, for clouds over ocean, to achieve the desired effect. This concentration exceeds the typical CDNC in marine environments by a factor of three. Since then, there have been several studies to determine the efficacy of MCB at a global level. Many other studies (e.g. [9–13]) assess the efficacy of MCB and discuss the albedo increase at different locations and under different climatic conditions. Korhonen et al. [14] were the first to show that accounting for the aerosol microphysics and transport in the atmosphere reduces the estimates of MBC efficacy. Beyond the aerosol indirect effect, Partanen et al. [15] and Jones & Haywood [16] also showed the significance of the direct radiative effect of injected aerosol particles.

Although the above-mentioned studies provide information on the global scale, the global models used are not able to simulate the emissions and transport of aerosols, and their interaction with the clouds in the cloud-resolving scale due to low spatial resolution used in the models. In most of the studies mentioned above, a uniform CDNC field over the geoengineered regions has been assumed which, in reality, would be very difficult to achieve with the proposed method [14,15]. This may have important implications for the size of the spray vessel fleet required to achieve the desired effect. Hence, smaller scale modelling is needed to resolve these issues. For example, on a cloud-resolving scale, Wang et al. [17] used a large eddy simulation (LES) model to study the microphysical aspects of MCB. They found that cloud seeding is less efficient in strongly precipitating, polluted and water vapour-limited regimes, and efficient only in weakly precipitating regions where injected aerosols suppress drizzle formation. Under dry or polluted conditions, only a 0.02 increase in albedo was obtained with the emission rate of \(1.45 \times 10^6\, \text{m}^{-2}\, \text{s}^{-1}\). Further, it was argued that a 0.06 increase is not even possible by increasing the emission rate threefold. However, one shortcoming in their model was that it did not account for the aerosol size distribution and the sensitivity of the result to aerosol composition. Recently, Jenkins et al. [18,19] performed LESs with a more realistic aerosol size distribution representation and found that aerosol injections are more efficient at night when boundary layer mixing is faster than during the day. Further, they found that water evaporating from sea spray causes cooling through latent heat uptake, which will lead to negative buoyancy. This cooling was found to be \(0.3\, \text{K}\) at maximum with the water evaporating in less than \(2\, \text{s}\). As a result, aerosol transport through the boundary layer was delayed and sedimentation to the sea surface was enhanced. However, in their model the water was added in the cloud water phase without explicit representation of aerosol water,
causing too rapid evaporation. In reality, water evaporation from particles continues as long as
the plume is mixed with drier air from the environment. In the studies by Wang et al. [17] and
Jenkins et al. [18], a 300 m model resolution was used. However, if the size of the emission source
is much less than the model resolution, the small-scale turbulence and particle dispersion right
after emission are not well resolved, which may lead to a severe underestimation of particle loss
rates. For instance, recently Stuart et al. [20] showed that, due to high particle concentrations near
the emission source, coagulation during and soon after the injection reduce the aerosol particle
number concentration by up to 50%. Injection rate is probably the overall main factor affecting
the particle concentration and should be taken into account when designing an actual vessel fleet.

In this work, we further study the aerosol–cloud interactions in MCB scenarios at a cloud-
resolving scale. We investigate how well aerosols are dispersed in the boundary layer before they
are removed through sedimentation. We use higher resolution than the previous cloud-resolving
studies in order to better resolve the details of the emission source point. For the first time, we
explicitly include the effect of water evaporation from sprayed particles and, thus, this study
serves as an evaluation of previous studies without the explicit treatment of aerosol water and
with fairly poor resolution.

2. Material and methods

In this study, we have used the large eddy model UCLALES [21,22] with the aerosol module
SALSA [23,24] to study aerosol–cloud interactions at a cloud-resolving scale. UCLALES has
been designed to study marine stratocumulus clouds and has been used in several previous
studies [25–28]. The turbulence closure uses the Smagorinsky model [29]. The sponge layer
at the top of the domain dampens the motion representing the free atmosphere and periodic
boundary conditions are used in horizontal directions. Momentum advection uses directionally
split fourth-order centred differences. The vertical advection is density weighted consistent
with the anelastic approximation. The radiation scheme is based on the Fu and Liou scheme
where the delta-fourstream method is used to solve the azimuth-averaged radiative transfer
equation [30]. Parametrization of rain and cloud water microphysics is based on the Seifert and
Beheng model [31].

With SALSA, we are able to realistically represent the aerosol size distributions found in the
marine environment. In SALSA, the whole spectrum of aerosols is divided into three subranges,
consisting of 10 size bins in total. Cloud droplet activation is calculated with the parametrization
by Abdul-Razzak & Ghan [32], so that the number concentration of cloud droplets formed at the
cloud base is a function of updraft velocity and the aerosol size distribution present in the air
parcel forming the cloud. In this study, the cloud droplet activation scheme was modified from
the default SALSA scheme presented by Kokkola et al. [23] by approximating the shape of the size
distribution assuming a linear dependency between the centres of two adjacent size bins [33]. We
do not consider cloud droplet activation taking place at the cloud top or edges.

To be able to compare our results with previous studies, we use the initial temperature and
moisture profiles for simulations from the first (RF01) DYCOMS II campaign flight [34]. The
surface temperature is set to 289 K and the wind speed is 5 m s⁻¹ in the east–west direction. The
large-scale wind divergence is given a value of 3.75 × 10⁻⁶ s⁻¹. The surface sensible heat flux and
surface latent heat flux are defined as 15.0 and 115.0 W m⁻², respectively.

We have used a domain size of 4.8 × 8.4 × 1.5 km with a 50 m horizontal and 10 m vertical
resolution in the boundary layer. Above 600 m, the vertical layer height increases at a ratio of
1.03, making a total of 105 layers. We represent background aerosols using a bimodal lognormal
distribution with a number concentration of 100 and 50 cm⁻³ in Aitken (mean size 30 nm) and
accumulation (200 nm) modes, respectively. The sea spray vessel moves with a speed of 2 m s⁻¹ in
the south–north direction, i.e. perpendicular to the wind direction as suggested by Salter et al. [7].
The vessel emits particles of 300 nm dry diameter [14,15], 25 m above the sea surface [7], at a rate
of 15 kg s⁻¹ (half the rate suggested by Salter et al. [7]) for about 40 min and travels across the
domain (4.8 km) once. From this emission rate, assuming the salinity of seawater to be 35 g kg⁻¹
Table 1. Processes included in the different simulation test cases performed at 04.00 and 13.00.

<table>
<thead>
<tr>
<th>Simulations</th>
<th>Water</th>
<th>Coagulation</th>
<th>Deposition</th>
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<tbody>
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<td>S_REF</td>
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<tr>
<td>S_DRY</td>
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<td>S_WET</td>
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<td>S_COAG</td>
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<td>S_DEP</td>
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and NaCl being the sole solute, we derived a particle number flux of $1.72 \times 10^{16} \text{s}^{-1}$. The post-injection analysis is restricted to 5 h, because we are mainly interested in the aerosol microphysics and dispersion taking place soon after the emission. We have performed a 24 h baseline simulation as well as geoengineering simulations with emission starting at two different times of the day, i.e. at 04.00 and 13.00.

To assess the contribution to particle loss due to different microphysical processes, we performed a set of simulations which accounted for different combinations of coagulation, dry deposition and aerosol water evaporation (table 1). In particular, to study removal processes, we performed one ‘lossless’ reference simulation (S_REF) which omitted all three processes, one simulation that only accounted for deposition (S_DEP) and one that only accounted for coagulation (S_COAG). In order to study the effect of latent cooling, we further performed two simulations, one including aerosol water (S_WET) and one without (S_DRY). The set of simulations was repeated at two different times of day, with emission starting 04.00 and 13.00. Additionally, we performed simulations at 150 and 300 m horizontal resolutions for S_WET and S_DRY.

3. Results and discussion

In all of the simulations, the aerosols injected at a height of 25 m are dispersed evenly in the whole simulation domain within approximately 4 h. Figure 1 shows the evolution of the aerosol plume at a height of 25 m in simulation S_DRY. The diagonal orientation resulting from the perpendicular vessel and wind speeds can be clearly seen (figure 1a,b). The nearly homogeneous distribution near the end of the simulation period is shown in figure 1c. Without aerosol water evaporation (in the S_DRY case), the particle number concentration loss during the emission due to surface deposition and coagulation was 7.45%, of which deposition contributed 3.24% (S_REF-S_COAG) and coagulation 4.11% (S_REF-S_DEP). After 5 h into the simulation, the particle number concentration decreased by 19.5%.

(a) Cooling effect due to water evaporation

The water content of the emitted sea salt particles adjusts to the ambient relative humidity (RH) within seconds after the emission. If the ambient water vapour pressure is below the vapour pressure over the emitted seawater droplet, the water will evaporate from emitted particles until equilibrium is reached. Apart from the resulting increase in water vapour content, the air is also cooled due to latent heat uptake, which decreases the saturation water vapour pressure. Both of these effects increase the ambient RH. Considering the small size (300 nm dry diameter; 1.2 µm wet diameter at emission) of the emitted sea salt particles, this means that equilibrium is reached within seconds after emission. The maximum possible cooling and resulting RH after injection but before mixing in ideal conditions can be calculated from mass and energy balance equations. In figure 2 we show both the cooling (figure 2a,b) and adjusted RH (figure 2c,d) caused by water evaporation at two different temperatures as a function of initial ambient RH and amount of injected seawater. What can be seen is that the temperature decrease due to latent heat uptake can
Figure 1. Aerosol concentration (cm$^{-3}$) at 25 m altitude: (a) 30, (b) 40 min and (c) 4 h after the initiation of emission. Wind blows at 5 m s$^{-1}$ from left to right and the vessel is moving upwards at 2 m s$^{-1}$. (Online version in colour.)

Figure 2. Effect of aerosol water evaporation on equilibrium temperature difference (K) and RH (%) as a function of emitted water amount and ambient RH at the initial temperatures of 285 K (a,c) and 295 K (b,d). (Online version in colour.)
be several degrees in dry conditions. In a typical marine boundary layer covered by stratocumulus cloud, the RH close to the surface is between approximately 70% and 90%, and in such conditions the maximum temperature decrease is limited to be less than 3 K. However, that is enough to cause negative buoyancy and changes in boundary layer dynamics, and it is much more than the temperature decrease suggested by Jenkins & Forster [19]. Evaporation of water from particles causes a temperature decrease close to and downwind from the emission source point. In our simulation S_WET, the seawater emission rate of 15 kg s$^{-1}$ from the vessel moving at a speed of 2 m s$^{-1}$ is causing an increase in total water content of approximately 6 g m$^{-3}$, and based on our theoretical estimate presented in figure 2 this is enough to cause a maximum temperature decrease of 2.1 K. However, owing to the dispersion of particles, the simulated temperature decrease is less than the maximum of 1.4 K close to the emission source (figure 3b). This is still much more than the 0.3 K found by Jenkins & Forster [19]. At the same time, the water evaporation also increases the water vapour content, and together with the temperature decrease this causes a rise in the RH to up to 97%. The RH gradually decreases to the background value in less than 1 min downwind from emissions. Jenkins & Forster [19] found that water evaporates within 2 s after emission, but when aerosol water is explicitly modelled the evaporative cooling will continue as long as the emitted plume is mixing with the dryer environment. In figure 3d, it can be clearly seen how the temperature decreases causing the negative buoyancy leading to a downdraft. This downdraft will form a pool of cold air with enhanced aerosol concentration close to the surface, leading to a delay in dispersion and more efficient deposition on the surface. Thus, the aerosol is reaching the cloud layer later than in the case of dry particles. This delay can be seen in figure 4a where a comparison of mean aerosol concentration has been made between the S_WET and S_DRY simulations at 400–850 m altitudes. The particle number concentration has a much higher peak close to the surface (figure 4c) when the cold pool formation due to latent heat uptake is simulated in the S_WET case. This is causing
a delay in the transport to higher altitude (figure 4a,b). However, the length of delay is estimated to be around 10 min and thus the particle loss during the emission period is only increased by a factor of 1.14.

The lifetime of the cold pool close to the surface was found to be very short in the night-time case. We can expect the lifetime to be affected both by the emission rate and by the boundary layer dynamics. A larger emission would mean more widespread cooling and vice versa. However, the emission rate used in our simulations is already high, so more interesting is the role of the boundary layer dynamics. We study this by changing the time of emission to be at 13.00, when the radiative cooling from the cloud top is not causing negative buoyancy and thus mixing of the boundary layer is slower. During the night-time emission, the mean vertical velocity variance is 0.22 m$^2$ s$^{-2}$ and at 13.00 it is 0.10 m$^2$ s$^{-2}$. Smaller variance leads to a stronger accumulation of particles close to the surface, as can be seen in figure 4, where dashed lines show the particle concentration after emission at different altitudes in daytime emission. The dispersion of particles into the cloud layer is delayed by approximately 20 min in S_WET compared with S_DRY (figure 4c). Overall, with aerosol water evaporation included, for daytime emissions, the particle loss during the emission period is increased by a factor of 1.26 compared with emission at night.

(b) Effect of resolution

In order to facilitate a better comparison with previous studies, we repeated the 04.00 simulations using 150 and 300 m horizontal resolution (figure 5). The clear difference in the simulations is that the temperature decrease due to water evaporation from particles is much smaller with 150 m (0.7 K) resolution (figure 5e) and even less with 300 m (0.4 K) resolution (figure 5f) when compared
with the 50 m case (1.4 K), as the average concentration of emitted seawater particles is lower with coarser resolution. This will lead to faster dispersion of particles into the whole boundary layer depth and less efficient particle removal. At the same time, as the maximum particle concentration is lower, the coagulation rate is lower as well. With a 50 m resolution, a loss of 7.45% of emitted particles was observed during the emission, while with 150 and 300 m resolutions the fraction of scavenged particles is 5.4% and 3.1%, respectively. Although there is a clear difference in particle loss rate due to resolution, the removal is still much slower than that reported by Stuart et al. [20], who used a very high-resolution plume model. Thus, when planning practical experiments, care should be taken in the design of the emission source to avoid too fast coagulation through high particle concentrations. Cloud fraction, liquid water content and CDNC are similar with different model resolution (not shown) during the 04.00 emission. Thus, in that respect, our results support the previous studies (at 300 m resolution) done by Wang et al. [17] and Jenkins & Forster [19].

4. Conclusion

It has been proposed that, to counteract the warming caused by increased GHG concentrations, the albedo of marine stratocumulus clouds could be enhanced by artificial injections of sea salt particles. The efficacy of this technique, commonly known as MCB, has been studied by many researchers using global-scale models, but studies with explicit aerosol microphysics on cloud-resolving scale are still rare. In our study, we use a high-resolution LES model coupled with aerosol module SALSA to study the behaviour of aerosols at the microphysical level before complete dispersion in the boundary layer. We model the particle emissions from a single moving source and study how aerosol microphysics including the explicit treatment of aerosol water is affecting the total particle number concentration and dispersion to the cloud layer and how the model resolution is affecting the results.

The dispersion of particles in the boundary layer is quick after the emission, and thus the plume is diluted before substantial coagulation loss. We find that, with an injection rate of 15 kg s$^{-1}$ of seawater during the 40 min emission, 7.45% of particles are scavenged by coagulation.
and surface deposition when we assume the particle dry size to be 300 nm. It is shown that the effect is enhanced when evaporation of water from sea spray is taken into account. Evaporation may cool the air by several degrees causing negative buoyancy, which leads to the formation of a cold pool of air close to the surface containing a high concentration of emitted particles. The extent and lifetime of the cold pool is dependent on the amount of emitted sea spray, boundary layer dynamics and meteorology. In the simulation, we found the maximum temperature decrease to be 1.4 K, leading to a 10–20 min delay in particle dispersion depending on the timing of the injection. Because of the enhancement of particle concentration close to the surface, 2.5–4.6% more particles are scavenged compared with simulations omitting aerosol water. More particles (4.6%) are scavenged during daytime emissions, when the boundary layer is more stable as there is no radiation cooling at the top of the cloud. As a decrease in ambient temperature so close to the sea surface would certainly affect the surface heat fluxes there, we performed an additional simulation to quantify the effect that our assumption of fixed surface heat fluxes has on the results. By keeping the sea surface temperature fixed and letting the heat fluxes vary according to the temperature differences near the surface, we found that the maximum change in sensible heat flux averaged over a $500 \times 1000$ m area is approximately $1.5 \text{ W m}^{-2}$. Considering the total sensible heat flux of $15 \text{ W m}^{-2}$ and the short time (several minutes) that the cold pool exists, we conclude that our assumption has no notable effect on the dispersion of aerosols in the boundary layer.

Altogether, in all simulations we find that over 80% of the particles are still prevailing 5 h after the emissions. The higher the resolution, the more persistent is the pool of cold air close to the surface and the more particles are scavenged due to both coagulation and sedimentation. It can be assumed that a further increase in resolution will capture the evolution of plume better within a few seconds after emission when the coagulation is most efficient and can lead to a rapid decrease in particle number [20], and it will be an issue of experimental design to avoid such high concentrations. Our results support previous findings [14,17,19] that earlier global studies may have overestimated the cloud concentration nuclei (CCN) enhancement; however, based on our results, the overestimation is not likely to be high enough to invalidate the findings. Note that in this study, in order to allow for the inclusion of the aerosol model SALSA, a relatively small domain size was chosen for the simulations, which makes it difficult to quantify the influence that the cold pools may have on the larger scale boundary layer dynamics. We further suggest that future studies related to marine stratocumulus brightening should be directed to regional studies using high enough resolution to capture single vessels emitting particles but still able to capture the dynamics of cloud systems over several weeks and several injection periods of new particles.

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