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ACPD 15, C4064–C4070, 2015

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Interactive comment on "Parameterizations for convective transport in various cloud-topped boundary layers" by M. Sikma and H. G. Ouwersloot

M. Sikma and H. G. Ouwersloot

martin.sikma@wur.nl

Received and published: 25 June 2015

We thank Referee #1 for his/her comments on the manuscript that will improve the paper and are glad that he/she agrees that the results are of interest to the community. We will respond to his/her comments point by point. The reviewer's comments are shown in italic.

General comments:



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1. the 'old' school (and the Reviewer belongs to this) believes it is much more accurate and physically consistent to estimate the mass flux directly, e.g. as function of the surface buoyancy flux), instead of estimating it as a product of two fitted quantities. This requires further analysis

The reviewer is right that it could be debated which method is more suitable to represent the convective transport of moisture. We therefore do not advocate that all models should use this procedure to calculate moisture transport this way. However, we do want to mention the possibility.

On the other hand, the convective transport of atmospheric tracers other than moisture, has to be calculated as a product of different functions. This is treated by, e.g. Ouwersloot et al. (2015). This clarification will be processed in the introduction of the revised manuscript.

2. the manuscript is a bit 'thin' in novelty and the authors should make clear what is actually new

To calculate the transport of atmospheric reactants, it is essential that the kinematic mass flux and chemical concentrations of the transported air are known. For the first time, we investigate the parameterization for the latter for 24 chemical species, over a wide range of conditions. To our knowledge, such a study has not been conducted before. Furthermore, the kinematic mass flux could be calculated directly by a convection routine, but this approach does not always have to be used. For example, in mixed-layer modelling the kinematic mass flux is calculated as described in the manuscript (see e.g., Neggers et al., 2006; van Stratum et al., 2014). Essential for these calculations, the current parameterization for active cloud area fraction is shown to be lacking and a novel, improved representation is introduced. Moreover, as the effectiveness of

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convective transport of atmospheric tracers is affected by the area of venting (Ouwersloot et al., 2015), this updated parameterization for active cloud area fraction is even important for tracer transport if mass fluxes are directly calculated by a convection routine.

We will emphasize the novelty of the study by highlighting the applications of and advancements in the parameterizations in the conclusions chapter.

3. the discussion of the parametrization needs and status in large-scale models is in parts a bit superficial also it is not clear how useful and for whom is in practice the relation for the core fraction of the species: eg in forecasting using a mass flux scheme these values are estimated from lifting parcels from near the surface with a certain excess values applying some strong entrainment

We answered this questions already partly in the question above, but the presented parameterizations are essential to deal with the transport of atmospheric compounds, other than water. As such, it is very relevant for models that take into account chemical transport. For instance, such a parameterization has recently been applied to the atmospheric chemistry - climate model EMAC (Ouwersloot et al., 2015) to accurately simulate transport of atmospheric compounds. We will update the manuscript to clarify the usefulness of the parameterization.

Specific comments:

-page 3, line 12 :('50-200 km)' NWP and GCMs run nowadays at 10-200 km globally and 1-2 km regionally

This is fixed in the revised manuscript.

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-page 3, lines 15-25: revise. Adjustment schemes do not transport mass as such, but adjust (relax) the thermodynamic profiles toward a moist adiabat. Please revise references herein as this is all quite inaccurate and obsolete including what you say about diffusive transport. You might have a look in the document below which summarizes also how mass flux schemes work in NWP and how tracer transport is done and also what are adjustment schemes and useful references http://old.ecmwf.int/newsevents/training/lecture_notes/LN_PA.html___("Atmospheric

http://old.ecmwf.int/newsevents/training/lecture_notes/LN_PA.html ("Atmospheric moist convection")

The reviewer raises a valid point. We re-checked the references and changed the paragraph into: "The impact of convective transport on atmospheric state variables (e.g., moisture and temperature), can be parameterized in large-scale models by using a convective adjustment scheme (e.g. Betts, 1986), an eddy-diffusion scheme (e.g. Soares et al., 2004), or the mass flux approach (e.g., Bechtold et al., 2001; Bretherton et al. 2003). In this study, we mainly focus on the latter, which also allows for convective transport of chemical compounds."

-page 4, line 15;'In contradiction' there is no contradiction, use different wording

The sentence is changed into: "By not applying the simplifications present in previous literature (e.g. Neggers et al., 2006), we developed a general formulation..."

-page 6, Eq (2): A mass flux should always include the factor rho (density) and have units kg/(m2 s)

We agree that for the general mass flux this is the case. However, since we are focusing

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on the kinematic mass flux, no factor rho is present in the formulas. We acknowledge that at some places the word "kinematic" was omitted, which is rectified in the revised manuscript.

-page 13, lines 5-8 and page 16 lines 6-7: you give references and say 'global models that use the parametrization of ... overestimate the mass transport'. None of these models computes mass transport using cloud fraction but directly estimates the mass flux! wrong references/literature for that problem

The reviewer is right. To our regret, we made a mistake here. The cloud area fraction parameterization of Cuijpers and Bechtold (1995) was not used for the computation of the mass flux in these models. We will change and clarify this in the revised manuscript.

-page 14, eq (12): This formulation can produce negative values in principle, robust?

That is right. When the mass flux is that strong that $\pm 85\%$ (1/1.18) of a grid cell is drained, even negative concentrations would result. In the application of convective tracer transport, one should take this in consideration. To prevent the unrealistically low concentrations, numerical solutions need to be applied. A first, simple solution would be to limit the total transport to never yield negative concentrations. More fitting solutions would be to e.g., introduce intermediate time steps for the convective tracer transport calculations or to account for the concentration evolution during a time step, as is applied by Ouwersloot et al. (2015).

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Interactive comment on Atmos. Chem. Phys. Discuss., 15, 10709, 2015.

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ACPD 15, C4059–C4063, 2015

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Interactive comment on "Parameterizations for convective transport in various cloud-topped boundary layers" by M. Sikma and H. G. Ouwersloot

M. Sikma and H. G. Ouwersloot

martin.sikma@wur.nl

Received and published: 25 June 2015

We thank W. Angevine for his comments that clarify the manuscript and increase the quality of the results. Below, we will respond on his comments point by point and include the changes that will be included in the revised manuscript. The reviewer's comments are shown in italic.

General comments:



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1. The abbreviation SCu is used for shallow cumulus. This is easy to confuse with stratocumulus. Maybe ShCu would be a better choice, or shallow cumulus could be spelled out each time.

We agree and replaced SCu by ShCu throughout the manuscript.

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2. p.10719 line 22 and following paragraph: It is not clear to me what is being claimed here or its relevance to the rest of the paper. Are you claiming a universal relationship between cloud and core area? Should this not be affected by changes in lapse rate, surface flux partitioning, etc., at least in some extreme cases? Furthermore, what does this ratio have to do with the rest of the paper? If such a relationship holds, why do you use two different functional forms for the cloud and core parameterizations?

With the rough relationship shown in Fig. 2b, we want to stress that not all clouds effectively transport air. Based on their characteristics, only approximately half of the clouds can be considered as active. Indeed, the exact ratio is case specific, but does not seem to deviate significantly between different cases. Therefore, this ratio (2.12) is presented to give a first impression about the relative impact. To only consider the clouds that enable vertical exchange, we further characterize the various types and present and quantify the area fraction of only the active clouds. In the revised manuscript we will update this paragraph to convey the message more clearly that a_c and a_{cc} are not similar and roughly differ by a factor of 2 during the phase with active convection. We will stress that the exact factor differs between conditions and that the independent parameterization of a_{cc} will be derived in Sect. 3.2.

3. There should be more attention to uncertainty and significance of the results. Some

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of the coefficients are compared to very slightly different values in the literature, but without information about, for example, the uncertainty of the fits used to derive them.

The reviewer raises a good point. By revising and developing the parameterizations, we used a least square error fit to fit the data most accurately. This least square error was not presented in the original document, but will be introduced in the figures and their discussion in the revised manuscript.

Specific comments:

1. The first sentence of the abstract suggests a more general study than what is presented. It might be better to say something like, "We investigate the representation of transport of atmospheric compounds by boundary-layer clouds..."

We agree with the reviewer's suggestion to be more specific and changed the sentence to: "We investigate the representation of convective transport of atmospheric compounds by boundary-layer clouds that can be applied in large-scale models."

2. p.10714 lines 2-4: It is not clear that forced clouds produce no transport. They can be quite deep in some cases, and may detrain. You should simply say that you neglect them here.

Regarding the forced clouds we use the cloud classification scheme of Stull (1985). We will clarify this earlier in the paragraph. According to Stull, forced clouds do not reach the level of free convection, which normally makes them quite shallow. However, additionally we will clarify the paragraph and state that we neglect forced clouds in this

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study.

3. p.10719 line 13: Please clarify the sentence. The number of clouds decreases, the total area stays constant, so the area of each cloud must increase. Right?

We thank the reviewer for pointing out this imprecise sentence. Instead of 'amount of forced clouds' we should have used 'area fraction of forced clouds'. Additionally, we should clarify that while a_{cc} remains constant, a_c decreases due to a decrease in the area of forced clouds, leading to a relative increase in the active cloud area fraction compared to the total cloud area fraction.

We changed the sentence to: "As this transport of energy out of the sub-cloud layer affects the thermal structures, the area fraction of forced clouds decreases due to a decrease in the amount of thermals that reach the cloud layer. The area fraction of active clouds is not significantly affected by this process, while a_c decreases, so that the $\frac{a_{cc}}{a_c}$ ratio increases."

4. p.10721 line 25: I don't understand what this has to do with an overestimate of cloud fraction. Cloud fraction must always be greater than or equal to core fraction, regardless of how well they are estimated. Please clarify.

The reviewer is right that this sentence is out of place. Since it does not contribute to the message, we will remove it from the document.

5. p.10724 line 9: It should be kept in mind (of the authors and readers) that the effects of segregation are usually quite small and depend on the reaction and mixing time scales. Are the effects significant here?

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We agree with the reviewer that the effects of chemical segregation are usually small. Shown by Ouwersloot et al. (2011) for clear sky conditions, deviations due to segregation are around the order of 12% for isoprene, which is within the uncertainty range of the measurements. However, in the case of cloud-topped boundary layers, the dynamical segregation can be substantial, as indicated in Fig. 5 of Ouwersloot et al. (2013). This is the background of why $\phi_{cc} \neq < \phi >$ and Eq. (12) is applied. Because the properties of escaping air differ from the mean, we have to take this effect into account. For chemistry, the effect is not quantified in this study, but will likely be relatively small due to compensating effects, as indicated by Ouwersloot et al. (2011).

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van Stratum, B. J. H., Vilà-Guerau de Arellano, J., van Heerwaarden, C. C. & Ouwersloot, H. G., *Subcloud-Layer Feedbacks Driven by the Mass Flux of Shallow Cumulus Convection over Land*, J. Atmos. Sci., 2014, 71, 881-895

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Interactive comment on Atmos. Chem. Phys. Discuss., 15, 10709, 2015.

List of relevant changes made in the manuscript.

General

Throughout the manuscript we made small modifications to clarify the message. At certain parts, we introduced sentences to convey the novelty, as suggested by the referees.

Introduction

Shifted one paragraph upwards to clarify the text. Small clarifications throughout the text.

Methodology

Clarified Sect. 2.1

Results

Sect. 3.1

End of section 3.1 is rewritten to clarify the purpose of the selection procedure.

Sect. 3.2

Introduced uncertainties in the parameters of the parameterization by using a covariance-matrix. Also a standard error to visualize the spread around the parameterization is introduced. The end of section 3.2 is rewritten and clarified, as we agreed with Referee #1 that the references were not correct regarding the message we wanted to convey.

Conclusion

The conclusion is clarified and more emphasis is laid on the novelty.

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Parameterizations for convective transport in various cloud-topped boundary layers

M. Sikma^{1,2} and H. G. Ouwersloot¹

¹Max Planck Institute for Chemistry, Mainz, Germany ²Meteorology and Air Quality Section, Wageningen University, Wageningen, the Netherlands

Correspondence to: M. Sikma (martin.sikma@wur.nl)

Abstract

We investigate the representation of convective transport of atmospheric compounds that can be applied in large-scale models by boundary-layer clouds. We focus on three key parameterizations that, when combined, express this transport: the area fraction of transporting clouds, the upward velocity in the cloud cores and the chemical concentrations at the cloud base. The first two parameterizations combined represent the kinematic mass flux by clouds.

To investigate the key parameterizations under a wide range of conditions, we use Large-Eddy Simulation model data for 10 meteorological situations, characterized by either shallow cumulus or stratocumulus clouds. The parameterizations are not tested for such a large data set before. In the analysis we show that the parameterization of the area fraction of clouds, currently used in mixed-layer models, is affected by boundary-layer dynamics. Therefore, we (i) simplify the independent variable used for the this parameterization, Q_1 , by considering the variability in moisture rather than in the saturation deficit . We show that there is an unambiguous dependence of the area fraction of clouds on the simplified Q_1 , and update the parameters in the parameterization to account for this simplification. We (ii) further demonstrate that the independent variable has to be evaluated locally to capture cloud presence. Furthermore, we (iii) show that the area fraction of transporting clouds is not represented by the parameterization for the total cloud area fraction, as is currently applied in large-scale models assumed in literature. To capture cloud transport, a novel active cloud area fraction parameterization is proposed.

Subsequently, the scaling of the upward velocity in the clouds' core by the Deardorff convective velocity scale and the parameterization for the concentration of atmospheric reactants at cloud base from literature are verified and improved by analyzing 6 SCu shallow cumulus cases. For the latter, we additionally discuss how the parameterization is affected by wind conditions. This study contributes to a more accurate estimation of convective transportin large-scale models, which occurs there at sub-grid scale.

Convective transport by shallow cumulus (SCuShCu) clouds is a key process in the lower atmosphere, as it regulates the partitioning of surface fluxes (Vilà-Guerau de Arellano et al., 2014; Lohou and Patton, 2014) and the temporal evolution of chemical reactants (Vilà-Guerau de Arellano et al., 2005; Ouwersloot et al., 2013). By venting air from the atmospheric boundary layer (ABL) to the free troposphere, SCu-ShCu strongly influence the ABL evolution, temperature, moisture content, and the variability of chemical species (Sorooshian et al., 2007; van Stratum et al., 2014). Besides their local effects, SCu ShCu contribute strongly to the spread in the estimation of climate sensitivities by affecting both longwave (greenhouse warming) and shortwave (reflective cooling) radiation (Boucher et al., 2013). This makes it essential to represent SCu ShCu and their effects accurately in atmospheric chemistry, climate and weather prediction models. However, due to the relatively coarse resolution of these models ($\sim 50 \sim 10-200 \text{ km}$), SCu (~ 1 globally), ShCu ($\sim 0.5-1 \text{ km}$) need to be treated as a sub-grid phenomena and are therefore required to be parameterized.

Convective transport The impact of convective transport on atmospheric state variables (e.g., moisture and temperature) can be parameterized in large-scale models by using a convective adjustment scheme (e.g. Betts, 1986), an eddy-diffusion scheme (e.g. Tiedtke et al., 1988) (e.g. Soares et al., 2004) or the mass flux approach (e.g. Bretherton et al., 2003) (e.g. Bechtold et al., 2001; Bretherton et al., 2003) . In this study, we mainly focus on the latter, which also allows for convective transport of chemical compounds. The mass flux approach is based on the mass continuity equation, where the mass flux is defined as the difference between the lateral entrainment and detrainment rate. By analyzing 10 numerical experiments performed by Large-Eddy Simulations (LES), we investigate three key parameterizations that can be used to represent mass transport in large-scale models, namely: the area fraction of clouds, the upward velocity in the cloud cores and the concentrations at the cloud base. The latter is also applicable when a convective adjustment or eddy-diffusion scheme is employed.

As the initiation of SCu ShCu formation depends on surface forcings the surface forcing and the thermodynamic state of the ABL, we discriminate between two situations: (i) the marine ABL, and (ii) the continental ABL. Since the formation of SCu-ShCu in the marine ABL is characterized by almost constant surface forcings a nearly constant surface forcing, resulting in steady-state conditions, this situation has been extensively studied (e.g., Neggers et al., 2004; de Rooy and Siebesma, 2008; Suselj et al., 2013). The marine steady-state SCu-ShCu case used in this study is the Barbados Oceanographic and Meteorological Experiment (BOMEX; Holland and Rasmusson, 1973). On the other hand, the continental ABL is affected by a diurnal cycle in surface forcings the surface forcing. The large variation in surface forcings forcing during day drive the initiation of SCu-ShCu formation, therefore impacting the dynamical structures in the ABL (Horn et al., 2015). As this situation is harder to study and therefore less investigated, four continental campaigns are selected, ranging from the mid-latitudes to the tropics, to serve as inspiration for the LES numerical experiments: the Tropical Forest and Fire Emissions Experiment (TROFFEE: Karl et al., 2007), the Gulf of Mexico Atmospheric Composition and Climate Study (Go-MACCS; Jiang et al., 2008), the Small Cumulus Microphysics Study (SCMS; Neggers et al., 2003) and the Atmospheric Radiation Measurements (ARM; Brown et al., 2002).

In this work, we simplify the statistical cloud area fraction parameterization as described by Cuijpers and Bechtold (1995, hereafter CB95) by considering the variability in moisture rather than the saturation deficit. In contradiction to simplifications proposed in literature (e.g. Chaboureau and Bechtold, 2002; Neggers et al., 2006) By not applying the simplifications present in previous literature (e.g. Neggers et al., 2006), we developed a general formulation that shows an unambiguous dependency of the cloud area fraction on the independent variable, Q_1 , for a wide range of thermodynamic conditions. For this, we perform 10 distinct numerical simulations, where we first focus on deriving a consistent representation for the total SCu ShCu cover. Furthermore, the assumption made by Neggers et al. (2006, hereafter NG06), entailing that the cloud area fraction parameterization can be used for the representation of the area fraction of active clouds, was recently shown not to be valid for a tropical (TROFFEE) case (Sikma et al., 2014). Here, we build on this finding by proposing a novel parameterization for the area fraction of active clouds, which is appropriate for convective transport. Subsequently, extending the work of Ouwersloot et al. (2013) and van Stratum et al. (2014), we present improvements on the scaling of the vertical convective velocity. As a result, we are able to accurately describe the mass flux in <u>SCu cloudsShCu</u>. We finalize by showing that the parameterization for concentrations of chemical species concentrations at cloud baseof Ouwersloot et al. (2013), as described by Ouwersloot et al. (2013), can be used under a wide range of conditions, although dynamical segregation slightly influences the results. Our findings can be used in large scale models to represent sub-grid scale convective transport, or in conceptual models to investigate <u>SCu</u> interactions (e.g., Vilà-Guerau de Arellano et al., 2012; van Stratum et al., 2014). Furthermore, as the the vertical velocity and cloud cover are essential to calculate cloud micro-physics and radiation properly (Arakawa, 2004), our results will enhance the representations of these in global models.

As shown by Ouwersloot et al. (2011), the chemical variability in clear sky conditions is affected by ABL dynamics, creating regions of high- and low concentrations, thereby modifying the mean reactivity. Since <u>SCu-ShCu</u> impact the dynamical structures in the ABL (Horn et al., 2015), it will enhance this segregation of species (Kim et al., 2004). As below the <u>SCuShCu</u>, the concentrations of chemical species differ more from cloud-layer concentrations than the mean concentrations in the ABL (Ouwersloot et al., 2013). Our findings can be used in large-scale models to represent sub-grid scale convective transport, or in conceptual models to investigate ShCu interactions (e.g., Vilà-Guerau de Arellano et al., 2012; van Stratum et al., 2014). Furthermore, as the vertical velocity and cloud cover are essential to calculate cloud micro-physics and radiation feedbacks properly (Arakawa, 2004), our results enhance the representation of these in global models.

The next section introduces the theory of mass flux and is followed by the descriptions of the model and numerical experiments. In the results, we first explore the effects of cloud venting on the temporal evolution of SCuShCu. This is followed by parameterizations of

the area fraction of <u>SCu-ShCu</u> venting and a scaling of the vertical convective velocity. We finalize with a validation and adjustment of the parameterization for the concentrations of chemical species at cloud base. While doing so, we discuss the role of dynamical segregation in the ABL.

2 Methodology

2.1 Cloud types and cloud distinction

To increase the representation of SCu and its effects in large-scale models, parameterizations are developed. As not all clouds transport ABL air to the free troposphere, it is necessary to Following Stull (1985), we discriminate between different cloud types for convective transport predictions. Since forced cloudsare, as not all clouds transport ABL air towards the free troposphere. Forced clouds, related to air parcels that reach the lifting condensation level, but are buoyantly too weak to reach the level of free convection, no net mass transport occurs by these clouds. Consequently, forced clouds are neglected in this study. Clouds that reach the level of free convection are marked as active clouds, since the release of latent heat increases the buoyancy of the cloudas the latent heat release increases the in-cloud buoyancy, thereby enhancing cloud growth. These active clouds transport mass, which affects the underlying ABL. When these As a result, they affect the underlying atmosphere by venting. When the active clouds decouple from the ABL thermals, they lose their supply of energy and become passive-clouds that. As a result, they do not contribute to the mass transfer anymore (Stull, 1985; Siebesma et al., 2003).

The part of the domain in which convective transport occurs, is quantified by the area fraction of clouds, which is defined at each level independently (Siebesma et al., 2003; Ouwersloot et al., 2013). Note that we cannot use cloud cover, as this property is not locally determined but based on the vertically integrated liquid water path. Furthermore, we distinguish in the remaining of the paper between all clouds and cloud cores, i.e. active

clouds, with subscript _c and _{cc}, respectively. As a result, we can distinguish four indicators for cloud presence, namely: cloud cover (c_c), cloud core cover (c_{cc}), area fraction of clouds (a_c) and area fraction of cloud cores (a_{cc}).

2.2 Mass flux parameterization

Mass transport can be approximated as the <u>kinematic</u> mass flux (M) multiplied with the spatial difference in the concentrations of chemical species at cloud base (ϕ) (Betts, 1973):

$$\overline{w'\phi'} = M\left(\phi_{cc} - \overline{\phi}(z_b)\right),\tag{1}$$

where ϕ_{cc} indicates the value in the cloud core, and $\overline{\phi}(z_b)$ indicates the domain averaged value at cloud base.

The kinematic mass flux, M, is defined by the area fraction of cloud cores (a_{cc}), the difference between the cloud core vertical velocity (w_{cc}) and the domain averaged vertical velocity at cloud base ($\overline{w}(z_b)$) (Betts, 1973), through

$$M = a_{\rm cc} (w_{\rm cc} - \overline{w}(z_{\rm b})). \tag{2}$$

For models that run on a coarser grid resolution than the width of a cloud core, the variables of Eq. (2) cannot be resolved explicitly and therefore need to be parameterized. We start by parameterizing the area fraction of cloud cores (a_{cc}). NG06 approximated a_{cc} by the total area fraction of clouds (a_c). The parameterization of a_c is developed by CB95, which uses

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(3)

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locally taken variables depending on temperature and moisture, and is expressed by

$$a_{c} = 0.5 + \beta \arctan(\gamma \cdot Q_{1}).$$

Here the constant $\beta = 0.36$ and $\gamma = 1.55$ represent a fit through the LES results of CB95. Q_1 is calculated as

$$Q_1 = \frac{\overline{s}}{\sigma_{\rm s}}.$$
(4)

Here, *s* denotes the saturation deficit and σ_s indicates the SD standard deviation of *s*. Lenderink and Siebesma (2000) assumed for simplicity that Q_1 can be represented as

$$Q_2 = \frac{q_{\rm t} - q_{\rm s}}{\sigma_q},\tag{5}$$

where q_t and q_s are, respectively, the total and saturation specific humidity, and σ_q is the spatial SD standard deviation of the specific humidity. Based on this work, NG06 applied this expression for a_{cc} , while the Q_2 is replaced by Q_3 , which is further simplified to be applicable in a mixed-layer slab model, according to

$$Q_3 = \frac{\langle q_{\rm t} \rangle - q_{\rm s}|_{\rm h}}{\sigma_{\rm q}|_{\rm h}}.$$
(6)

Here, $\langle q_t \rangle$ is the total specific humidity averaged over the mixed layer and $q_s|_h$ and $\sigma_q|_h$ represents the respective values at the mixed-layer top. Although these adapted variables indeed coincidentally converted the expression for a_c to a reasonable prediction for a_{cc} for the case evaluated by NG06, we demonstrate in Sect. 3.2 that this is not valid for all thermodynamic and dynamic conditions and that a different formulation should be applied.

As shown by Neggers et al. (2004), the cloud core vertical velocity can be scaled with the Deardorff convective velocity scale (w_*) (Deardorff, 1970). Building on this work,

Ouwersloot et al. (2013) and van Stratum et al. (2014) showed for several ShCu cases that the inclusion of a prefactor improved this scaling:

 $w_{\rm cc} \approx 0.84 w_*$,

which further will be verified -extended in this study. Furthermore, Ouwersloot et al. (2013) showed that as shown by Ouwersloot et al. (2013), the concentrations of chemical species at the base of the active clouds can be parameterized as:

$$\phi_{ extsf{cc}} - \overline{\phi}(z_{ extsf{b}}) pprox -1.23 \left(\overline{\phi}(z_{ extsf{b}}) - \langle \phi
angle
ight) \,.$$

2.3 LES model

The numerical model used in this study is DALES 4.0. This version contains several improvements over version 3.2 (Heus et al., 2010), including additional elements (e.g. new landsurface submodels Vilà-Guerau de Arellano et al., 2014) and the introduction of an anelastic approximation for density changes with height (Boing et al., 2014). In DALES, most ($\sim 90\%$) of the turbulent processes are solved explicitly in a convective ABL when run on a grid resolution of 100 m or less. As a result, only parameterizations for the smaller scale turbulent structures are needed, which makes it an adequate tool to use in our study. With the use of the Boussinesg approximation, the filtered Navier-Stokes equation is solved (Heus et al., 2010). Furthermore, DALES consists of no-slip boundary conditions at the bottom and periodic boundary conditions at the sides. At the top of the domain, a sponge layer is located present which damps fluctuations caused by e.g. convection waves.

2.4 Numerical experiments

In all cases, the horizontal grid resolution is set to $50 \,\mathrm{m} \times 50 \,\mathrm{m}$, which covers an area of $12 \,\text{km} \times 12 \,\text{km}$. A larger domain or increase in grid resolution proved not to be of significance. The vertical resolution and extent are case dependent and are listed in Table 1.

(8)

The direction of the wind is always set in the *x* direction (*u* component), but differences are present in the velocities (Table 1). Also the case dependent surface kinematic heat and moisture fluxes are prescribed. Furthermore, the ABL top is defined at the height where the gradient of the virtual potential temperature (θ_v) exceeds 50 % of the maximum gradient in the vertical profile of θ_v (Ouwersloot et al., 2011).

Ten numerical experiments are run to simulate a range of SGu-ShCu and stratocumulus cases. Regarding the SGuShCu, 5 situations (TROFFEE, GoMACCS, SCMS, ARM, BOMEX) are selected. Additionally, TROFFEE+ and SCMS- consider an adapted wind velocity compared to the original TROFFEE and SCMS cases, respectively. The SCMS_{cold} case represents an adaptation on SCMS, where the initial vertical profile of θ is lowered by 2 K. This is done to represent a transition from stratocumulus to shallow cumulus, as discussed in Sect. 3.2. Regarding the stratocumulus, 2 situations (ATEX and DYCOMS-II) are analyzed.

To investigate the diurnal impact of SCu convection, the SCu The ShCu simulations start in the early morning and are based on daytime convective conditions. Depending on the geophysical location, the UV The radiation is calculated as a function of time, depending on the geophysical location. The chemical mechanism applied in the SCu ShCu cases is identical as described in Ouwersloot et al. (2013) and contains 20 reactant species and three passive tracers. The latter are an emitted tracer (INERT; emission of 1 ppb m s⁻¹), an inert species that is initially only present in the ABL (BLS) and an inert species that is initially present in the free troposphere (FTS). To ensure that the reactions are fully resolved, the time step is forced to a maximum of 1 s. For all cases, the data is stored at a 1 min interval.

The stratocumulus experiments are solely performed to include representative data for the upper regime of the total cloud area fraction parameterization. Therefore, no chemical scheme is applied.

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3 Results and discussion

3.1 Temporal evolution of shallow cumulus

The temporal evolution of the total and active cloud area fraction is presented in Fig. 1. The cases TROFFEE and ARM are clearly affected by a different partitioning in sensible and latent heat fluxes, caused by the diurnal cycle in incoming solar radiation. This demonstrates that the initiation of SCu formation is dependent on the surface forcing. As a result, the SCu-ShCu start to develop from mid-morning and diminish in the late afternoon. The GoMACCS and SCMS cases show different dynamics compared to TROFFEE or ARM, as SCu-ShCu start to develop in the early morning (06:30 and 07:00 LT, respectively). This can be explained by a high relative humidity in the initial profiles at the start of the day, therefore favoring cloud formation (not shown). The reason for these high values can be found in the geophysical location of these cases, which are close to the ocean, even though they are classified as continental cases. In contrast to the continental numerical experiments, the BOMEX case is characterized by an almost a nearly constant surface forcing over the ocean and is therefore classified as a marine steady-state case. In the first half an hour, moisture and heat is building up in the ABL, which causes the sudden formation of SCu ShCu around 05:30 LT. After 08:00 LT, the transport of energy is proportional to the supply of energy from the surface fluxes, so the temporal evolution of the area fraction of clouds and cores is in steady-state.

As is visible in Fig. 1, for all continental SCu cases all continental ShCu cases show a time lag of one hour is present in the initiation of a_{cc} compared to a_c . This can be explained by forced clouds, which are dominant during the first hour. This is also visible in Fig. 2a, where the ratio between a_c and a_{cc} is shown. By focusing on the forced phase, it is visible that the area fraction of clouds increases during time, but that almost no active clouds are present. It is interesting to note that the dynamics in the BOMEX case are not comparable with the other cases, since it does not start in this forced phase, as mentioned earlier. In the next phase, the transition phase, the amount area fraction of clouds remains roughly equal, but the forced clouds are replaced by active clouds. During this process, the a_{cc} increases fast,

indicating that the threshold for active SCu–ShCu growth is overcome. In the end of the transition phase, cloud venting affects the sub-cloud layer structures by redistributing the thermals (Horn et al., 2015). As this transport of energy out of the sub-cloud layer affects the thermal structures, the amount area fraction of forced clouds decreases, due to a decrease in the amount of thermals that reach the cloud layer. Since the a_{cc} The area fraction of active clouds is not significantly affected by this process, the area fraction of active clouds relatively while a_c decreases, so that the $\frac{a_{cc}}{a_c}$ ration increases. This process is clearly visible in the ARM and GoMACCS case (Fig. 1). When the transport of energy is proportional to the increase in energy by the surface fluxes, we identify this period as the active phase. During the active phase, the ratio between a_c and a_{cc} is roughly constant ($a_c = 2.12a_{cc}$), while both gradually decrease in time. In the final phase, the dissolving phase, the number of active clouds reduces rapidly due to the diminished surface forcings, so that the clouds become decoupled forcing. In other words, the clouds decouple from the boundary-layer thermals and active clouds increases (see Fig. 2a).

As we are mainly interested in mass transport by SCu, we select for further analysis our data such that we capture To only consider the clouds that enable vertical exchange, we perform a selection procedure based on the time period where SCu transport is dominant. In when the presence of a_{cc} is high. We show in Fig. 2b, it is visible that during the active phase the ratio between that during this time period, a_c and a_{cc} is not dependent on the atmospheric conditions which differ between the numerical experiments. An increase in surface forcing directly translates into an increase in cloud venting, as the mass transfer and surface fluxes are tightly coupled. An increase in surface fluxes would cause both are coupled, but a_c and a_{cc} to increase, with a_c increasing decreases faster by a factor of 2.12 (d = 0.93). Here, d represents the index of agreement (Wilmott, 1981). Comparing our unfiltered results with the results from van Stratum et al. (2014) , we find that we have a slightly lower but similar slope of 2.35 (d = 0.89) for our unfiltered results instead of This rough relationship is a valid first approximation, but one should note that the exact factor differs between conditions and that an independent parameterization of both components is needed, which will be derived in Sect. 3.2. To show the importance of this selection procedure, we compare our (selected) data with the data (no selection) from van Stratum et al. (2014). Their relationship of 2.46 (d = 0.77) - In is higher than ours, indicating that the effects of mass transport are underestimated, which decreases the accuracy of the mass transport parameterizations. Therefore, in the remainder of this paper, we use the selected data to evaluate the parameterizations and scalings scaling for ShCu transport.

3.2 Parameterizing the area fraction

To asses the validity of the simplified statistical cloud area fraction parameterization (hereafter a_c-parameterization) of NG06 (Eq. 12 therein) under different thermodynamic conditions, ten numerical experiments are run-performed to simulate a wide range of SCu-ShCu and stratocumulus cases (see Sect. 2.4). As is shown in Fig. 3a, the a_c -parameterization of NG06 is not able to consistently represent the total cloud area fraction. Furthermore, by using Q_3 , no clear dependency is visible of the cloud area fraction on specific moisture conditions. An explanation for this could be found in the dependency of Q_3 (Eq. 6) to the volume of the ABL and the thickness of the transition zone, i.e. the region between cloud base and ABL. This dependency is only introduced in NG06, since Q_1 in CB95 and Q_2 are evaluated locally. Although NG06 simplified the expression for a_c with help of Lenderink and Siebesma (2000) to reproduce the occurrence of active clouds with an atmospheric mixed-layer model, we show that this simplification can introduce significant errors depending on the evaluated case. Therefore, a revision of the a_c -parameterization is needed such that the cloud area fraction can be reproduced for a wide range of atmospheric conditions. Furthermore, an independent representation of the active cloud area fraction, necessary for convective transport, is needed. In these analyses, we use the locally determined Q_2 (Eq. 5) as indicator.

To include as many different a wide range of boundary-layer physics and cloud conditions between the SCu-ShCu and stratocumulus cases (i.e., between $Q_2 = -2$ and 1)as possible, two, two additional transition simulations, ATEX and SCMS_{cold}, are shown. ATEX represents a case where SCu-ShCu convection starts to develop, but an inversion causes

(9)

the build up of moisture near the ABL top, resulting in a stratocumulus layer. Another approach is used for the SCMS_{cold} simulation, where the initial vertical profiles of θ were decreased by 2 K. As a result, the relative humidity is close to 100% near ABL top in the morning, thereby creating a stratocumulus layer. When the surface fluxes start to increase, the stratocumulus layer breaks and convection starts to occur and the stratocumulus layer breaks. As is visible in Fig. 3b, a typical SCu-ShCu situation is present in the late afternoon which is comparable with the other SCu-ShCu cases and is captured by the revised a_c -parameterization. As is shown in Fig. 3b, using the proper index variable, Q_2 , results in a well-defined dependence of cloud area fraction. Furthermore, using this approach we can deduce an accurate parameterization for a_c for all numerical experiments. By using a least square fitting method the Levenberg-Marquardt algorithm for least square curve fitting, we find

 $a_{\rm c} = 0.5 + 0.34 \pm \alpha$ $\arctan(1.85\beta Q_2 + 2.33\gamma)$.

In where $\alpha = 0.34$ (0.002), $\beta = 1.85$ (0.063) and $\gamma = 2.33$ (0.111). In brackets, the standard error of the parameter estimate is shown, which is calculated with use of a covariance-matrix over the parameters. The residual standard error yields 0.036, which is calculated via the reduced chi-squared method. In ATEX and SCMS_{cold}, both the SCu-ShCu and stratocumulus regimes are generally captured well by Eq. (9), while only the transition between this regime remains troublesome. This deviation is reflected in the relatively large residual standard error. Focusing solely on a cloud fraction lower than $a_c = 0.3$, i.e. ShCu cases, the residual standard error yields 0.007, as also shown in Fig. 4.

As mentioned before, a_{cc} cannot be parameterized by the expression for a_c . This is confirmed by the $a_c = 2.12 a_{cc}$ relation of Fig. 2. shown in Fig. 2. In Fig. 4, we show that a separate parameterization is needed for a_{cc} . Inspired by Eq. (7), we derive We derive

 $a_{\rm cc} = 0.292 \, Q_2^{-2} \, \underline{}, \tag{10}$

where the standard error in the parameter yields 0.001. The residual error yields 0.005. Next to a_{cc} , we display the a_c data (shaded) in Fig. 4, together with its parame-

terization, in order to demonstrate that a_{c} and a_{cc} can be well-represented independently, but that these representations are not similar. As such, using the prediction of $a_{\rm c}$ for to predict $a_{\rm cc}$, as is currently assumed in literature (e.g. Neggers et al., 2006), will lead to wrong predictions of the active cloud area fraction. Global models that use the parameterization of CB95 or Chaboureau and Bechtold (2002) 2004), FGCM-0 (Fushan et al., 2005), or Can/ overestimate the mass transport. This finding is consistent overestimation of boundary layer clouds in a single column model a tropical case (Suselj et al., 2013) . Furthermore, as explained earlier, the simplification Furthermore, the simplified a_c-parameterization of NG06 for use in an atmospheric mixed layer model introduces inconsistencies depending on the evaluated case. Using Eqs. (9) and (10) removes this dependency. Therefore, it is important to use the novel parameterization of these inconsistencies and is therefore essential to predict in-cloud transport and associated feedbacks correctly. Besides an improved representation of in-cloud transport in mixed-layer models, Eq. (10) to increase the accuracy of in-cloudtransport and associated feedbacks.Note that in large-scale models the local SD of moisture is needed to diagnose Q₂, which can be represented using the formulations of Tompkins (2002) is also relevant for global models that deal with the transport of atmospheric compounds other than water (e.g. the EMAC atmospheric chemistry-climate model (Ouwersloot et al., 2015)), as the area fraction of active clouds is essential to calculate the correct transport outwards a grid cell.

3.3 Scaling of convective transport

As the cloud core vertical velocity, w_{cc} , is the final component of the kinematic mass flux formulation (Eq. 2), we evaluate the scaling of Neggers et al. (2004) for various atmospheric conditions to complete the kinematic mass flux parameterization. Neggers et al. (2004) showed that the w_{cc} can be scaled with the Deardorff convective velocity scale (w_*). Building on this work, Ouwersloot et al. (2013) and van Stratum et al. (2014) found that a prefactor of 0.84 improved this scaling. Their analysis was based on four SCu Shou cases, where

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no filtering was applied selection on the data was applied to distinguish between active SCu ShCu and forced/passive SCuShCu. Therefore, the presence of forced and passive clouds disturbs the scaling of w_{cc} . As a result, their value of the scaling is lower due to the weaker vertical velocities related to forced clouds. By only taking the active phase into account, we find the following relation (Fig. 5):

 $w_{\rm cc} = 0.91 \, w_*,$ (11)

with d = 0.90. The high index of agreement shows that this relation is not affected much by different boundary-layer dynamics and structures. However, as is visible in Fig. 5, the TROFFEE case is less well represented by the scaling. If we apply a fit through TROFFEE data alone, we find a scaling factor of 0.845 (d = 0.74), which is comparable to the result of Ouwersloot et al. (2013) who found 0.84 (d = 0.94). The deviation of this case compared to other cases could be explained by a relative deep ABL depth ($\sim 2 \text{ km}$). Combined with strong surface forcings a strong surface forcing, the w_* increases strongly, while the w_{cc} is not significantly affected. This results in a lower scaling constant.

3.4 Parameterizing reactant transport

In this section we focus on the final component of the expression for convective transport of atmospheric compounds (Eq. 1), namely the concentration of chemical species at cloud base($_{c}(\phi_{cc} - \overline{\phi}(z_b))$). The parameterizationis, proposed by Ouwersloot et al. (2013)who, showed that the concentrations of chemical species at the base of active SCu ShCu can be predicted by Eq. (8) for a tropical case (TROFFEE). However, they stress that their parameterization can be influenced by ABL dynamics ABL dynamics could influence the parameterization. Therefore, we test the parameterization for all continental SCu ShCu cases. The relation is illustrated for four chemical species (i.e., INERT, BLS, isoprene and CO) in Fig. 6- In total, but the least squares regression through the concentrations of is fit

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through all 24 evaluated chemical species yields for our continental SCu data. This yields:

$$\phi_{cc} - \overline{\phi}(z_b) \approx -1.18 \left(\overline{\phi}(z_b) - \langle \phi \rangle \right)$$
 (12)

For all relations, the amount of data yields a *d* of index of agreement is 1.00.

Comparing these results with Ouwersloot et al. (2013) shows that We find similar results as Ouwersloot et al. (2013) but our constant is slightly less negative than their -1.23. Since we use the least squares method to find the optimum scaling constant, it implies means that compounds with the largest differences between $(\phi(z_b) - \langle \phi \rangle)$ affect the scaling constant the most. As is shown in Fig. 6, this means that INERT has a dominating influence. Focusing on this compound (inset), we see find that wind tends to increase the differences between species in the cloud core compared to their average at cloud base, while for a free convection situation the opposite is visible. As a result, the closure constant of Eq. (12) shifts slightly. In this case, we find This results in a slope of -1.17 in case of wind and a slope of -1.19 in case of free convection (not shown). We identify for the INERT species that dynamical segregation is occurring in the ABL, as shown for INERT in Fig. 7 and discussed by Ouwersloot et al. (2013) for a tropical case. Rising motions in the ABL transport high concentration of the emitted species upwards, while lower concentration of INERT are found in the downward motions. Therefore, higher concentrations of species are transported towards the free troposphere by cloud venting as would be expected compared to a well-mixed situation. However the effects of chemical segregation are usually small for clear sky situations, it could be substantial for cloud-topped boundary layers due to cloud venting. As a result, the chemical parameterizations and scalings are effected. As Furthermore, as is shown in Fig. 7, wind affects the distance and upward velocities in the thermals, resulting in less, but wider thermals in our domain. Furthermore, This affects the vertical transport of species is slower and decreases this transport (max. $3.5 \,\mathrm{m\,s^{-1}}$) compared to a free convection situation (max. $5.0 \,\mathrm{m\,s^{-1}}$) where the thermals are more narrow. As an effect, the transport of chemical species to the cloud layer is less in the wind case, resulting in a smaller difference between ϕ_{cc} and $\overline{\phi}(z_{b})$, which decreases the magnitude of the scaling constant of Eq. (12). Next to affecting the an effect on convective transport, one has to note that dynam-

ical segregation also modifies the mean reactivity in the ABL, as was shown by Ouwersloot et al. (2011) for clear sky conditions and Kim et al. (2004) in SCu ShCu situations.

4 Conclusions

In this paper, the The representation of sub-grid convective transport of atmospheric compounds in large-scale models by boundary-layer clouds is investigated. We focused on three key parameterizations that express this transport, namely: the area fraction of clouds, the upward velocity in the cloud cores and the concentrations at cloud base. The parameterizations are investigated under a wide range of conditions with the use of Large-Eddy Simulation (LES) model data from seven boundary-layer cloud cases, ranging from SCu shallow cumulus (partly cloud cover) to stratocumulus (totally overcast). Next to the seven standard boundary-layer cloud cases, three additional cases are simulated that are slightly adapted to provide additional information needed for deriving the parameterizations.

We found that the simplified statistical cloud area fraction parameterization, and the combined variables it uses as input, are influenced by the structure of the atmospheric boundary layer (ABL). Therefore, the parameterization was not applicable to a wide range of conditions. We simplified and updated this parameterization by considering the variability in moisture rather than the saturation deficit, and show that this parameterization has to be evaluated locally to properly accurately capture cloud presence. Furthermore, we demonstrate that the parameterization for the total cloud area fraction cannot be used to represent the to area fraction of active clouds, as is currently assumed in literatureand applied in large-scale models. This leads to an overestimation of the in-cloud mass transport when this parameterization is used. To capture this cloud transport ,-properly, we propose a novel parameterizationis proposed. Besides its usefulness in mixed-layer models, it is also relevant for global models to capture the area fraction of a grid cell in which chemicals are drained to upper layers.

Moreover, we evaluated the scaling of the cloud core vertical velocity with the Deardorff convective velocity scale by using 6 continental representative SCu ShCu cases. It was

found that the previously published relation holds, but that a higher closure constant improves this relation. Combining the parameterizations for the area fraction of active clouds and the cloud core vertical velocity, we are able to accurately represent the kinematic mass flux induced by SCu clouds ShCu clouds, applicable over a wide range of conditions.

To finalize our analysis, the parameterization of reactant concentrations at the base of active clouds is investigated for the was investigated for 6 continental SCu cases. ShCu cases, as in previous literature it was only validated for a tropical case. We found a minor spread in the derived closure constants for the parameterization, depending on whether a background wind is was present or not, which can be explained by the affected dynamical segregation of chemical species in the ABL. However, this spread is was small and a general derived closure constant can be applied for parameterizations in large-scale models. In total, we validated and updated three robust parameterizations that can be used in large-scale models to represent sub-grid scale convective transport of chemical species atmospheric compounds and propose a novel parameterization essential for ShCu venting.

Acknowledgements. The authors would like to thank Jordi Vilà-Guerau de Arellano for the insightful discussions and comments. We also thank W. Angevine and an anonymous reviewer who helped improve the quality of this work. The numerical simulations were performed with the supercomputer facilities at SURFsara and sponsored by the project NCF-NWO SH-060-13.

The article processing charges for this open-access publication were covered by the Max Planck Society.

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Case	Vertical resolution [m]	Vertical extent [m]	Wind $u \text{ comp.}$ [m s ⁻¹]	Case type	Reference to LES case/ Comments
TROFFEE	20	5990	0.0	Continental ShCu	Ouwersloot et al. (2013)
TROFFEE+	20	5990	5.0	Continental ShCu	Adapted, including wind
GoMACCS	25	4988	0.0	Continental ShCu	Jiang et al. (2008)
SCMS	20	3990	5.65685ª	Continental ShCu	Neggers et al. (2003)
SCMS-	20	3990	0.0	Continental ShCu	Adapted, removing wind
ARM	20	4490	10.0	Continental ShCu	Brown et al. (2002)
BOMEX	40	3180	-8.75 ^b	Marine ShCu	Siebesma et al. (2003)
SCMS _{cold}	20	3990	5.65685ª	Transition case	Adapted, decrease of 2 K in θ
ATEX	20	3990	-8.0^{b}	Transition case	Stevens et al. (2001)
DYCOMS-II	10	1595	3.02 ^b	Marine stratocumulus	Stevens et al. (2005)

 Table 1. Experimental setup of the shallow cumulus and stratocumulus cases.

^a Rotated wind vector which is comparable with the actual SCMS case.

^b Height dependent wind profiles, starting at surface. A more detailed description can be found in the references.



0.25-

0.25

Figure 1. Temporal evolution of the domain averaged maximum area fraction (N) of clouds and cores for the <u>SCu-ShCu</u> cases (Table 1). The blue lines denote an experiment with wind (indicated with a "+"), while the red lines indicate a free convection case.



Figure 2. Scaling of the area fraction of clouds as a function of the area fraction of cloud cores. In (a) all data is presented, where a distinction is made between different phases of convection during day. The lines represent the best fit through the active phase and all data, forced through 0. In (b) the selected data is shown for each <u>Scu-ShCu</u> case. Circles indicate free convection situations, while crosses indicate wind situations. To differentiate BOMEX from the other cases, BOMEX is marked with triangles in (a) and (b). Furthermore, *d* represents the index of agreement.

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Figure 3. Area fraction of clouds as (a) a function of the normalized saturation deficit (Q_3 ; Eq. 6) as described in Neggers et al. (2006) and (b) as a function of the normalized saturation deficit at cloud base (Q_2 ; Eq. 5). Negative Q_2 and Q_3 values indicate SCu ShCu clouds, while positive values denote stratocumulus clouds. The dashed lines indicate the parameterizations based on Q_3 and Q_2 . SE represents the residual standard error.



Figure 4. The area fraction of cloud cores (a_{cc}) is represented by the coloured symbols, while the a_c for all <u>SCu-ShCu</u> cases is shown in grey. Both area fractions are shown as a function of the normalized saturation deficit at cloud base (Q_2) . Crosses denote a wind situations, while circles indicate free convection situations. <u>SE represents the residual standard error</u>. The lines represent the best fit parameterizations for a_c and a_{cc} .

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Figure 5. Scaling of the cloud core vertical velocity (w_{cc}) as a function of the Deardorff convective velocity scale (w_*). Circles represent free convection situations, crosses indicate wind situations. The line represents a least square fit, which is forced through 0. *d* represents the index of agreement.



Figure 6. Parameterization for $\phi_{cc} - \overline{\phi}(z_b)$ as a function of $\overline{\phi}(z_b) - \langle \phi \rangle$ proposed by Ouwersloot et al. (2013). Here, ϕ represents the 24 transported species (note that only INERT, BLS, isoprene and CO are shown). Circles represent free convecition situations, crosses indicate wind situations. The solid line represents a least square fit trough all data, which is forced through 0. *d* represents the index of agreement. The inset shows solely the INERT species for wind and no wind experiments.

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Figure 7. Vertical cross sections of INERT for the TROFFEE case for (a) a free convection situation and (b) a wind situation. The white arrows indicate wind vectors of the v and w component. In (b), the mean horizontal wind is substracted from the flow to identify the vertical patterns. The white horizontal line around 1400 m denotes the ABL height, which is calculated using the threshold gradient method. In black, contour lines are shown for w, starting at a lower limit of 2 m s^{-1} with intervals of 1.5 m s^{-1} .

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