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The scientific basis for a satellite mission to retrieve CCN concentrations and their impacts on convective clouds

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Abstract

The cloud -mediated radiative forcing is widely recognized as the main source of uncertainty in our knowledge of the anthropogenic climate forcing and in our understanding of climate sensitivity. Current outstanding challenges are (1) global measurements of cloud condensation nuclei (CCN) in the cloudy boundary layer from space, and, (2) disentangling the effects of aerosols from the thermodynamic and meteorological effects on the clouds. Here we present a new concept for a way to overcome these two challenges, using relatively simple passive satellite measurements in the visible and IR. The idea is to use the clouds themselves as natural CCN chambers by retrieving simultaneously the number of activated aerosols at cloud base, N_a , and the cloud base updraft speed. The N_a is obtained by analyzing the distribution of cloud drop effective radius in convective elements as a function distance above cloud base. The cloud base updraft velocities are estimated by double stereoscopic viewing and tracking of the evolution of cloud surface features just above cloud base. In order to resolve the vertical dimension of the clouds, the field of view will be 100 m for the microphysical retrievals, and 50 m for the stereoscopic measurements. The viewing geometry will be 30 degrees off nadir eastward, with the Sun in the back at 30 degrees off zenith westward, which requires a Sun synchronous orbit at 14:00 LST. Having measured simultaneously the thermodynamic environment, the vertical motions of the clouds, their microstructure and the CCN concentration will allow separating the dynamic from the CCN effects. This concept is being applied in the proposed satellite mission named Clouds, Hazards and Aerosols Survey for Earth Researchers (CHASER).

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1.1 Uncertainty in aerosol cloud-mediated forcing drives the climate uncertainty

The effects of aerosols on clouds are recognized by the IPCC (2007) as the largest sources of uncertainty in the quantification of anthropogenic perturbations to the Earth's energy balance. Therefore the reduction of this uncertainty is necessary for improving our understanding of the present climate and for the reduction of the uncertainties in the prediction of future climates (e.g. Andreae et al., 2005; Kiehl, 2007). The uncertainty in aerosol cloud-mediated radiative forcing is composed of two large and highly uncertain opposite effects from shallow and deep clouds (Rosenfeld et al., 2012b). This underlines the importance of conducting global measurements of the aerosol effects on clouds. This paper introduces a new concept for doing these kinds of measurements from a space, showing the feasibility of what has been considered impossible until now.

1.2 Ways by which aerosols affect cloud microphysical, precipitation and radiation properties

Aerosols are extremely important to cloud radiative forcing because cloud droplets are nucleated on pre-existing aerosols that serve as cloud condensation nuclei (CCN). The cloud droplet concentrations, N_d , that form near cloud base depend on the supersaturation (S) activation spectrum, $CCN(S)$, and on the cloud base updraft speed, w_b . The CCN that actually nucleate cloud droplets in specific clouds are referred to as activated CCN, and their number density before dilution by mixing with ambient non-cloudy air is denoted as N_a . The mixing with ambient air reduces N_a to N_d . This number determines the variability of the fundamental properties of the clouds at a given meteorological situation. The albedo of clouds with a given value of vertically-integrated liquid water path increases with N_d (Twomey, 1977). With greater N_d smaller droplets are formed,

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reducing coalescence and inhibiting rain production (Gunn and Phillips, 1957; Squires, 1958). The dynamic response to the rain suppression lengthens the life-time and increases the cloud cover of marine stratocumulus (Albrecht, 1989; Rosenfeld et al., 2006). In deep convective clouds rain forms eventually, but the aerosol-induced delay in its formation to greater heights was shown to cause cloud invigoration and additional electrification (Andreae et al., 2004; Rosenfeld et al., 2008). The aerosol-induced vertical growth and the consequent expansion of the anvils into cirrus inflicts large positive radiative forcing (Koren et al., 2010), in contrast to the strong negative forcing that is caused by the aerosol effect on shallow clouds (e.g. Albrecht, 1989). In the context of 5 tropical cyclones the invigoration of clouds at the periphery of the storms occurs at the expense of a reduction in the amount of air converging into the storm center, hence reducing the maximum wind speeds (Rosenfeld et al., 2012a). The aerosols were found even to affect the potential of deep convective clouds to produce large hail and tornadoes, where clouds with added pollution aerosols become potentially more damaging 10 (Rosenfeld and Bell, 2011). The impact of aerosols on cloud invigoration and the vertical distribution of latent heating can also modulate regional and large scale circulation systems, as observed by Bell et al. (2008).

1.3 Uncertainties in global measurements of cloud-mediated radiative forcing by aerosols

20 Based on this brief summary of aerosol effects on clouds, it is evident that having accurate measurements of CCN activation spectra as a function of water vapor supersaturation – $CCN(S)$, is vital for a quantitative understanding of the weather systems and the aerosol impacts on Earth energy budget and climate. This information is necessary for disentangling the effects of aerosols from those of the green house gases 25 on the Earth energy budget, and hence for understanding climate change and climate prediction.

Global measurements are practical only from space. Aerosol properties such as aerosol optical depth have been measured from space, but the interpretation of the

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measurable quantities into $CCN(S)$ is still challenging. The radiative forcing caused by impacts of aerosol on clouds is inferred from satellite observations where aerosol optical properties are compared to cloud radiative properties. This practice is inaccurate, mainly because of the poor ability to convert the aerosol optical depth (AOD)

5 into concentrations of cloud condensation nuclei (CCN) that actually interact with the clouds (Andreae, 2009). The inaccuracies are caused by the fact that aerosols are not measured at the same height as the cloud base, cloud contamination (i.e. clouds enhancing the aerosol signals), and aerosol swelling in the moist environment near the clouds. Inaccuracies in the cloud properties are caused by mixing clouds that feed

10 from the boundary layer with clouds that have elevated bases above the boundary layer, hence not interacting with the aerosols there. Another yet unresolved challenge is correct retrieval of the properties of ensembles of convective clouds. A serious limitation is the inability of the aerosols and clouds in conditions of high fractional cloud coverage to be retrieved simultaneously. As a result, the practice of relating retrieved

15 cloud to retrieved aerosol properties is plagued by large random errors in both parameters. Mathematically, adding random noise to two physically linearly related variables (e.g. $Y = aX$) causes the correlation to decrease and the slope to tend toward zero. Therefore, the large observational errors in the dependence of clouds on aerosol properties have likely caused an underestimate of the slopes of their relationships, which

20 is defined to be the observed aerosol indirect effect (AIE). This error might explain the low estimates of the observed AIE as compared to results of model simulations (e.g. Quass et al., 2009; Penner et al., 2011).

2 A new proposed concept for measuring AIE

This paper introduces a new concept for measuring simultaneously $CCN(S)$ and the microphysical and dynamical properties of the clouds for assessing quantitatively the cloud-aerosol interactions with global coverage from satellites. A proposed satellite mission named “Clouds, Hazards, and Aerosols Survey for Earth Researchers”

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(CHASER) is based on this concept (Renno et al., 2012). The satellite has two main instruments: the Multi Spectral Imager (MSI) that will have a field of view of 100 m by 102.4 km at the center of the ground track for all wavelengths. The satellite view will point 30° off nadir eastward with the Sun at 30° off zenith on the west, at local solar time of 14:00 LST. The MSI will have 12 spectral bands in the visible, near IR and thermal IR, as shown in Table 1.

Cloud vertical motions will be obtained by using a sequence of stereoscopic images that map the evolution of the cloud surface. The Multi Angle Imager (MAI) onboard CHASER has this capability. The MAI consists of three visible high resolution cameras. 10 The center camera points 30° off nadir eastward. The other two cameras point 30° off nadir across-track (eastward) and $\pm 30^\circ$ off nadir along-track with respect to the center camera to achieve a stereo geometry with the Sun at the back as illustrated in Fig. 1. The field of view of each camera is 50 m by 102.4 km, with a spatial resolution of 50 m at their ground center track. The three cameras cross scan the same ground track, 15 providing a sequence of three images 1 min apart, from three different viewing angles. The idea of vertical microphysical profiling from space was introduced by Rosenfeld and Lensky (1998), using the Advanced Very High Resolution Radiometer (AVHRR), observing the temperature and particle effective radius (r_e) at the tops of convective clouds at various stages of vertical development. This idea was improved to a proposed 20 side scanning satellite (Martins et al., 2011), and further developed with enhancing the resolution and with the addition of the MAI for CHASER (Renno et al., 2012).

Traditionally, CCN are measured in laboratory or field experiments by a cloud chamber where known water vapor supersaturation (S) is applied to an air sample and the activated cloud droplets are counted. Here we view clouds as natural cloud chambers where the number of nucleated aerosols into cloud droplets, N_a , and S can be retrieved, and hence the CCN spectrum can be obtained. Specifically, the approach uses the retrieved N_a from the dependence of the cloud drop size on the potential amount of condensed cloud water as a function of distance above cloud base, along 25 with retrieved updrafts near cloud base, w_b , for calculating the CCN activation spectra

as a function of S . This is used to map CCN in the cloudy boundary layer globally. This is important because this maps CCN and w_b , the fundamental physical entities determining N_a , which in turn determine the rest of the cloud properties (e.g. cloud drop effective radius and initiation of rain as a function of height above base). Retrieving N_a and cloud base updraft velocities is required for disentangling the impacts of dynamics from microphysics on the various cloud properties.

The concept of using ground-based cloud observations for retrieving CCN was originally proposed by Feingold et al. (1998). They retrieved CCN using measurements of w_b with vertically pointing Doppler cloud radar, and N_d estimated from a combination of 5 cloud radar and passive microwave measurements. Obtaining the size distribution of the aerosols by Lidar or other measurements can provide the CCN supersaturation activation spectrum. This method was applied to ground based measurements of marine 10 stratocumulus clouds at Point Reyes to obtain the aerosol effects on cloud radiative forcing (McCominsky and Feingold, 2011). They showed that the variability in liquid 15 water path (LWP) that occurs due to 3-D effects and cloud mixing tends to reduce the measured strength of the albedo effect.

The proposed methodology represents a significant advancement, as it can use 20 satellite measurements and therefore provide a global coverage of retrieved CCN and cloud properties. Furthermore, variability in LWP for a cloud with a given depth and N_d is not a source of error for this methodology.

3 N_a as a fundamental extensive cloud property

The number of activated aerosols at cloud base, N_a , is determined by the aerosol size distribution, chemical composition, and water vapor supersaturation at cloud base. The latter is determined by $CCN(S)$ and the updraft speed at cloud base. In a hypothetical 25 cloud parcel rising adiabatically, the diameters of the cloud droplets grow in proportion to the cube root of the adiabatic cloud liquid water content [mixing ratio], q_{La} . This occurs because $q_{La} = 4/3 \rho_L \pi r_v^3 N_a$, where r_v is the mean cloud droplet volume

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4 N_a controls the height for the onset of warm rain

The droplet condensational growth is determined by the number of activated CCN (N_a) and the height above cloud base, D . When the droplet effective radius (r_e) exceeds $\sim 14 \mu\text{m}$, considerable precipitation mass is likely to form in growing convective clouds 5 regardless of the extent of entrainment and mixing. This is because the low droplet concentrations and liquid water content in the diluted parts of the cloud, which slow the coalescence rate, is compensated by the extra time and potential spectrum broadening due to typically weaker updrafts and preferred (partial) evaporation of the smaller droplets upon mixing. Additionally, the rate of droplet coalescence is proportional to 10 $\sim r_e^5$ which practically implies the existence of a threshold r_e above which efficient warm rain formation can occur, and also because the vertical profile of r_e , even in diluted clouds, closely follows the theoretical adiabatic condensational growth curve. The small observed deviations are mainly caused by deviations from purely inhomogeneous mixing which cause partial droplet evaporation. Consequently, the minimal cloud 15 depth for precipitation formation (D_p) must theoretically change nearly linearly with N_a because the adiabatic water content increases nearly linearly with D_p , with a slope that depends on the cloud base parameters. This means that in highly polluted clouds or where strong cloud-base updrafts occur (i.e. high N_a), clouds may need to grow well 20 above the freezing level, even in tropical atmosphere, before precipitation forms either by warm or by mixed-phase processes. For full detail see Freud and Rosenfeld (2012).

5 Retrieving N_d from satellites

Satellite retrieval of the column number of cloud droplets [cm^{-2}] is based on the retrieved vertical liquid water path (LWP). When LWP is given, the vertically averaged cloud droplet number density N_d can be estimated. Indirect measurements of N_a by 25 satellite and lidar retrievals were previously applied to shallow marine stratiform clouds, with the main assumption that the clouds are composed of nearly adiabatic elements

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(Bennartz, 2007; Brenguier et al., 2000; Schuller et al., 2003; Snider et al., 2010). These retrievals had large uncertainties mainly due to violation of the adiabatic assumption and were not always validated with direct measurements. Furthermore, this methodology is not applicable to convective clouds due to their large departure from the assumption that they are close to adiabatic as compared to marine stratocumulus, and also due to the variable cloud top heights and depths at scales smaller than the typical satellite sensor resolution.

The fact that N_d is vertically averaged and is subject to an unknown extent of dilution with ambient air renders it to be related only loosely to CCN. In contrast, N_a is the fundamental property representing the combined effects of CCN(S) and cloud base updraft velocity, w_b , which in turn determine other cloud properties. Consequently, the objective of this study is to introduce a method by which satellite observations of clouds can be used for retrieving N_a and its precursors, CCN(S) and w_b .

6 The scientific basis for retrieving N_a

Recent studies based on airborne measurements in deep convective clouds show that the cloud droplet effective radius (r_e) anywhere in the cloud is a strong function of the number of activated CCN (N_a) and the distance above cloud base (D). The entrainment of sub-saturated air from the surroundings of the cloud and the mixing with the cloudy air, although reducing the cloud droplet concentrations and the liquid water content by evaporation and dilution, apparently do not have a strong effect on r_e . The common explanation is that the mixing is strongly inhomogeneous, i.e. that the cloud droplets at the edges of a blob of entrained sub-saturated air quickly and completely evaporate while increasing the vapor content in the entrained air, until it is saturated and cannot evaporate additional droplets. This lowers the droplet concentrations but leaves the shape of the droplet size spectra unchanged and hence does not affect r_e . This is why, if extreme inhomogeneous mixing is assumed and the cloud base properties are known, N_a can be derived from the vertical profile of r_e as long as the droplets grow

mainly by condensation. When droplet coalescence is active and warm rain forms, then the unique relation between the vertical profile of r_e and N_a weakens. Therefore the derivation of N_a is expected to be most accurate for r_e values smaller than $\sim 13\text{ }\mu\text{m}$. Errors in the measurements of r_e and departures from the extreme inhomogeneous assumption would affect the determination of N_a and therefore the effects of mixing would need to be accounted for.

Accurate measurements of r_e require vertical resolutions of the order of 100 m. Moreover, accurate measurements of r_e as a function of the vertical distance above cloud base, D , require the minimization of 3-dimensional effects. The reduction of this effect can be accomplished by making measurements of r_e at wavelengths of about 3.7 μm because the strong absorption of radiation by water at this wavelength reduces 3-dimensional effects (Rosenfeld et al., 2004; Painemal and Zuidema, 2011). At shorter wavelengths where absorption is weaker (1.6 and 2.1 μm), a larger fraction of the light exiting a measured cloud pixel has been scattered by cloud particles inside and outside of the intended cloud measurement volume. Therefore, measurements at 3.7 μm are inherently more accurate for retrieving N_a than measurements at 1.6 or 2.1 μm .

Taking into account the deviation from the extreme inhomogeneous mixing scenario when deriving N_a from the vertical profile of r_e is not straightforward. Freud et al. (2011) showed that with the inhomogeneous mixing assumption N_a was over-estimated on average by $\sim 30\text{ }\%$. This is because the measured r_e needs to be extrapolated to its adiabatic value (and converted to r_v) before Eq. (1) can be applied. How much to add to the measured r_e depends mainly on the amount of entrainment and the relative humidity (RH) of the entrained air. The RH may be estimated from the measurements of the absorption of the water vapor (in the 1120–1150 nm band) near the edges of the clouds. The vertical profile of vapor near clouds will be retrieved by the derivative of the precipitable water as a function of height, when measured above the cloudy pixels at different heights in the cloud cluster of interest. The relative humidity will be calculated based on the known temperature as a function of height, as obtained from

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the measured cloud top temperatures at the various heights. This is expected to be much more accurate than the alternatives of forecast/reanalysis or derived by satellite inversion techniques (e.g. Singh and Bhatia, 2006 and the references therein).

Here we retrieve N_a using the following algorithm:

- 5 1. Retrieve $r_e(D)$, the vertical profile of r_e as a function of vertical distance above cloud base.
2. Convert the satellite retrieved r_e to r_v , using the very tight and invariable linear relations between the two, as shown in Fig. 2 here and more fully Fig. 5 of Freud and Rosenfeld (2012).
- 10 3. Invoke the assumption of inhomogeneous mixing, which at the extreme means that r_v equals r_{va} , the adiabatic r_v . This allows retrieving N_a in the following way:

$$N_a = 3/(4q_{La}\rho_L\pi r_{va}^3) \quad (1)$$

- 15 4. Correct N_a for the extent of homogeneous mixing and the resultant reduction of r_v with respect to r_{va} . This is done based on the relative humidity near the cloud. This procedure will be tuned by analysis of already available aircraft measurements (Freud et al., 2011).

Instead of correcting for the extent of homogeneous mixing (step 4 above), it can be assumed that the highest r_e values from each level in the cloud (there will be a range of r_e values for each level because of the high resolution of the retrievals) represent the least diluted cloud elements, and therefore are close to the adiabatic r_e . Therefore Eq. (1) may be used for deriving N_a (after replacing r_e with r_v) from the fitting of the best r_e profile to the highest r_e values at each level. This profile would resemble the enveloping curve shown in Fig. 2 (dark grey), which was calculated for an assumed N_a . Charts like the one shown in Fig. 3 can be used for obtaining N_a when $r_e(D)$ is retrieved. 20 25 This way for deriving N_a does not require accounting for the mixing inhomogeneity, so it is quite simple and straight-forward. On the other hand it only utilizes a relatively small

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7 The scientific basis for obtaining the updraft velocity near cloud base

Cloud vertical motions can be obtained in principle by using a sequence of stereoscopic images that map the evolution of the cloud surface. The Multi Angle Imager (MAI) onboard CHASER has this capability. Two stereoscopic views 1 min apart can in principle provide the vertical component of the cloud surface evolution, by tracking protuberances at the cloud envelope. Tracking such protuberances near cloud base with an oblique view from the side can provide the vertical motion of these features, which approximates the mean updraft at cloud base. An illustration of such cloud features is given in Fig. 6. The challenge here is separating the observed movement into horizontal and vertical components. This can be assisted by the independent knowledge of the height for the central view of the cloud surface. This height can be obtained from the cloud temperature as measured by the MSI that is aligned with the center camera of the MAI. The cloud surface temperature can be uniquely related to cloud surface height in convective clouds, when using the moist adiabatic lapse rate and the cloud base temperature. Systematic bias errors in the height-temperature relations in the same cloud cluster would not affect the accuracy of the retrieval, and the vertical lapse rate is rather constant, as determined by known thermodynamic factors that do not vary much within the domain.

The accuracy of the height retrieval by two Multi-angle Imaging SpectroRadiometer (MISR) cameras is about 300–500 m (Hovarth and Davis, 2001; Moroney et al., 2002). The MISR has a horizontal nadir resolution of 275 m, versus the 50 m footprint of the MAI. This alone should bring down the error by a factor of 5, resulting in an accuracy of 60–100 m. The off nadir tilt of the MAI viewing geometry and the height reference provide additional significant improvement in the accuracy. Using a fixed reference height of a flat cloud base at the top of a well-mixed boundary layer for all three views can further improve the accuracy by a factor of about two, leading to an accuracy of about 50 m.

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8 Combining N_a and W_b for obtaining the CCN spectra

Earlier researchers used CCN spectra extensively because they can be obtained directly from in situ measurements with cloud chambers. Often, these CCN spectra were parameterized using a power law of the form $CCN(S) = CCN(1\%) \times S^b$, such that the spectra could be represented by a CCN concentration normalized to $S = 1\%$, and an exponent b . More recent studies have moved away from this approach for two reasons. First, it was found that the power law was often a poor approximation for the observed CCN spectra, and therefore, b -values could not be reliably associated with specific aerosol types (Gunthe et al., 2009; Rose et al., 2010). The second and more important reason is that this representation treats CCN as a “black box” by using a parameterization that does not contain any fundamental physical properties, and thus it is not amenable to prognostic modeling.

Current CCN research shows that the activation of an aerosol particle at a specific S is predominantly determined by the number of water-soluble molecules or ions that it contains. The number of soluble molecules or ions, in turn, depends on the aerosol volume and composition (the number of soluble molecules per unit volume). The volume of the aerosol particle is specified by its dry diameter while the composition is specified by the hygroscopicity parameter, κ (Petters and Kreidenweis, 2007). In this approach, the CCN properties of aerosols can be related directly to their basic physical and chemical properties, and therefore are amenable to prognostic modeling. State-of-the-art in situ measurements are based on size selection of aerosol particles followed by their activation to CCN in a cloud chamber, so that the dependence of activation on size and composition can be studied separately (Rose et al., 2008, Su et al., 2010).

These data are most beneficial for improving our understanding of how aerosols affect climate if they are used to test and improve the prognostic aerosol models that are incorporated into climate models. For this purpose, the $CCN(S)$ spectra obtained from the CHASER measurements of the concentration of activated CCN as a function of the water vapor supersaturation, $N_a(S)$, are considered to represent the CCN

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concentrations as a function of particle size (d) and composition (Reutter et al., 2009). Figure 7 illustrates the relation between S and N_a calculated as a function of the up-draft speed near the cloud base (w_b), which can be described by $S = 3.78 w_b^{0.64} N_a^{-0.4}$. These calculations indicate that overestimates of 20 % in N_a cause S to be underestimated by 7 %, whereas overestimates of 20 % in w_b cause S to be overestimated by 12 %. Since the error of the estimates of N_a and w_b is ~10 % to 20 %, the error in the calculation of S is ~10 %.

5 Since aerosol particle size and composition affect CCN activation and can vary independently of one another, there is no unique way to convert a $\text{CCN}(S)$ spectrum into a $\text{CCN}(d, S)$ spectrum that would allow the retrieval of the desired functions: $N_a(d)$ and $\kappa(d)$. That is, the particle number concentration and hygroscopicity cannot be retrieved as a function of size without additional assumptions. Fortunately, recent research has yielded considerable information about κ from both theoretical studies and laboratory and field measurements. This research shows that submicron aerosols, 10 which constitute essentially almost all CCN, can usually be represented as linear mixtures of an organic component with $\kappa \approx 0.1$ to 0.2 and an inorganic component with $\kappa \approx 0.6$ (Gunthe et al., 2009; Rose et al., 2011). Where appropriate, an additional sea salt component with a $\kappa \approx 1.3$ can be introduced. This approach has provided a global 15 model of κ distributions that is in very good agreement with observations (Pringle et al., 2010). A comparison with observations shows that modeled and observed κ agree to ≤ 0.05 for 10 out of 14 stations. Field campaigns have also shown that κ does not usually change strongly with size over the size range relevant to CCN active in convective 20 clouds (0.05–0.5 μm).

Once κ is known or assumed, the distribution $\text{CCN}(S)$ can be inverted to a distribution $\text{CCN}(d)$ because for each κ there is a functional relationship between the supersaturation and the critical particle diameter above which particles can be activated to cloud droplets. Thus, each supersaturation step in the $\text{CCN}(S)$ spectrum corresponds 25 to a size step in the $\text{CCN}(d)$ spectrum, and the difference in CCN number concentration between the supersaturation steps yields the number of CCN in the corresponding

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size interval (Su et al., 2010). An alternative approach would be to apply a model that predicts both aerosol size distribution and κ on the basis of on aerosol source/sink and transport processes, such as the Global Model of Aerosol Processes (GLOMAP) (Spracklen et al., 2005a, b, 2010) Although the size distributions have larger errors associated with them than κ has, both can be used as priors with specified weights and uncertainties, and an optimal κ and $\text{CCN}(d)$ can be derived from the $\text{CCN}(S)$ spectrum.

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9 Using the retrieved CCN and cloud properties for disentangling thermodynamics from aerosol effects

10 Marked contrasts in the behavior of moist convection has long been recognized. These various contrasts have been increasingly well quantified on a global basis by satellite observations over the last twenty years, and are summarized in Table 2. Large hail (Williams et al., 2005), lightning activity (Christian et al., 2003) and upper level cirrus cloud (Kent et al., 1995) are all substantially more prevalent over land than over ocean.

15 In contrast, the prevalence of warm rain showers, in which ice particles are absent, is found over oceans (Williams and Stanfill, 2002; Liu and Zipser, 2009).

The traditional physical explanations for these contrasts, also reported in Table 2, are generally based on thermodynamic differences between land and ocean, which in turn are linked to contrasts in the surface properties (Table 3) of crustal materials and 20 liquid water. The stronger heating of the land surface is responsible for greater air-surface temperature contrast and greater instability there (Williams and Renno, 1993), and for the larger updrafts at both cloud base height (Williams and Stanfill, 2002) and at higher levels of deep convection (Jorgenson and LeMone, 1989). The relatively recent upsurge of interest and observational attention to atmospheric aerosol (Molinié 25 and Pontikis, 1995; Rosenfeld and Woodley, 2003; Hicks et al., 2005) has led to alternative physical explanations for the land/ocean contrasts (Table 2) that depend on the regulation of cloud droplet size and compete with the thermodynamic ones, so strongly

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that the two effects are difficult to distinguish from each other (Williams et al., 2002; Williams and Stanfill, 2002). New satellite-based methods are needed to address both explanations.

The plausibility of two possible explanations is underscored by the firmly established contrast in cloud condensation nuclei concentrations between land and ocean that is found in many regions (Table 3). Oceanic air is often clean, while continental air is polluted in many areas (Schaefer and Day, 1998). This difference (Table 2) is widely attributed to the prevalence of aerosol sources over land (by smoke from fire, air pollution, emissions from vegetation, and lofting of mineral dust and by volcanic eruptions), and the weaker aerosol sources over the sea. The general contrast in CCN concentrations between polluted and clean regions is an order of magnitude (Andreae, 2009). But as certain as the contrast between mostly polluted land and clean ocean in CCN is the land/ocean contrast in cloud base height (Williams and Satori, 2004; Williams et al., 2005), a quantity entirely independent of CCN concentrations. The thickness of warm cloud (difference between freezing level height and cloud base height) is necessarily generally greater over ocean and favors coalescence and the removal of cloud water as warm rain. The thermodynamic effect of thicker warm cloud over oceans mimics the aerosol effect there of cleaner air, smaller CCN concentration, larger cloud droplets and increased coalescence. Over oceans, the surface sensible heat flux is smaller and so the cloud base updraft strength is smaller (Table 3), nucleating smaller fraction of the aerosols into cloud droplets and allowing more time for coalescence and again favoring warm rain (Table 2).

Considering now the other three phenomena in Table 2 – lightning, large hail and upper tropospheric cirrus cloud – all three depend on the supply of condensate to the upper cloud. Lightning is mediated by graupel colliding with ice crystals. This graupel forms from supercooled water that has survived the coalescence process. Large hail forms by accretion of supercooled water in strong updrafts. Finally, upper tropospheric cirrus cloud depends on the nucleation of ice particles in the mixed phase region that is promoted by strong continental updrafts and by aerosol-rich air.

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The disentanglement of thermodynamic and aerosol contributions to land/ocean contrasts in moist convection will require the simultaneous measurement of thermodynamic variables (CAPE, sensible heat flux, cloud base height, updraft speed) and the CCN at cloud base height, on the same set of clouds, comparing cloud properties over 5 clean and polluted continental and marine regions. Then sensitivity analyses can be performed based on partial derivatives with respect to key thermodynamic and aerosol variables to quantify the various contributions.

Compelling evidence for a role for anthropogenic aerosol in the competition with thermodynamics in modifying moist convection is found in recent findings on the “weekend effect” (Bell et al., 2009; Rosenfeld and Bell, 2011). In parallel with aerosol measurements showing a deficit concentration on weekends are findings that lightning, hail and tornadoes are all suppressed (by 10–20 %) over weekends relative to week days. In the absence of evidence for changes in thermodynamic quantities on the weekly time scale over continents, a distinct role for aerosol remains.

Thermodynamic explanations have been favored in explaining the dependence of lightning activity on island area (Williams et al., 2004), the semiannual variation of global lightning activity (Williams, 1994) and the interannual variation of lightning activity, both regionally (Hamid et al., 2001; Yoshida et al., 2007) and globally (Williams, 1992; Satori et al., 2009). Comprehensive studies of aerosol effects on lightning activity 15 on the interannual time scale are lacking.

More recent efforts to distinguish thermodynamic from aerosol contributions in explaining local variations in oceanic lightning activity, arriving at the conclusion that aerosol effects are dominant, can be found in Yuan et al. (2011).

10 Summary

25 The scientific basis for a satellite mission to retrieve CCN and investigate their impacts on convective clouds is introduced here. It addresses the outstanding problem of cloud-mediated aerosol impacts on cloud radiative and thermodynamic forcing. The approach measures simultaneously the cloud composition and thermodynamic

properties, and infers the number of activated CCN, N_a , from the vertical evolution of the retrieved cloud r_e in convective elements. The N_a can be retrieved with an accuracy of about 20 %. To achieve this, multispectral measurements of the sun-illuminated side of the clouds at a resolution of 100 m are planned by the proposed CHASER satellite mission (Renno et al., 2012).

The concentration of CCN can be obtained if the maximum vapor supersaturation at cloud base height, S , is known. This, in turn, can be obtained from cloud base updraft speed, which can be retrieved from very high resolution (50 m) dual stereoscopic images at oblique view of 30 degrees off nadir for the center camera. The analysis with the benefit of knowledge that cloud base is flat over a well mixed boundary layer can yield vertical motions w_b near cloud base with an accuracy of $0.2\text{--}0.5\text{ m s}^{-1}$. A bias error of 20 % in w_b causes S to be biased by 12 % in the same direction. A 20 % bias error in N_a causes S to be biased by 7 % in the opposite direction. This allows the retrieval of the supersaturation activation spectra of the CCN with an accuracy of 10–20 %. The hygroscopicity parameter of the aerosol, κ , can be obtained from emission sources and aerosol transport models at an accuracy of 0.1–0.2. This allows the calculation of the size spectra of the CCN aerosols, which is an essential aerosol property for simulating aerosol effects on cloud microstructure and precipitation forming processes.

The aerosol properties are retrieved from the cloudy boundary layer, where other satellite-based methods of direct aerosol measurements are blind. This method is also free of problems that have plagued direct aerosol measurements, such as swelling of the particles at high RH, cloud contamination, and aerosols that are not at the same level as the cloud base.

Retrieving the vertical microstructure and thermodynamic properties of the clouds as well as the horizontal and vertical cloud motions allows obtaining simultaneous CCN concentrations, dynamics and thermodynamics of the clouds. This will make it possible to address effectively the outstanding challenge of disentangling meteorology from the aerosol effects on clouds, and hence the aerosol radiative and thermodynamic climate forcing. Section 9 introduces an approach for doing that.

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This will make it possible to have a major improvement in the accuracy of aerosol indirect radiative forcing and reduce substantially the magnitude of our uncertainty, which presently dominates the overall uncertainty in the magnitude of anthropogenic climate forcing.

5 *Acknowledgements.* The idea of retrieving CCN from space was stimulated by the discussions at the working group of remote sensing cloud-aerosol-precipitation-climate interactions, sponsored by the International Space Science Institute in Bern, Switzerland. We thank NASA Glenn Research Center, the University of Michigan, and DLR for supporting the development of the CHASER Mission concept. Many engineers, managers and technical staff played important
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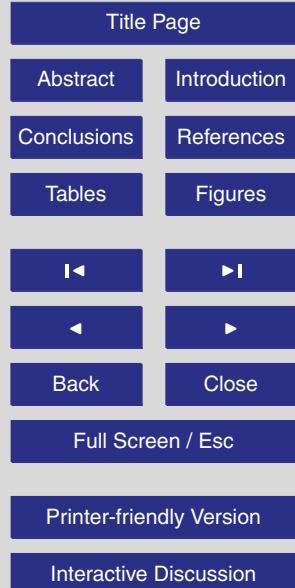


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Table 1. The spectral bands of the Multi-Spectral Imager, which will have a footprint of 100 m.

Wave band	Channel name
450–470 nm	Blue
560–580 nm	Green
670–690 nm	Red
1070–1200 nm	Vapor wide
1120–1150 nm	Vapor narrow
1365–1395 nm	Cirrus absorption
2080–2150 nm	2.1 Phase water
2200–2310 nm	2.3 Phase ice
3420–3950 nm	3.7 Effective radius
8.3–9.25 μm	8.7 Temperature
10.2–11.2 μm	10.7 Temperature
11.2–12.3 μm	11.7 Temperature



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Table 2. Contrast in land-ocean behavior in moist convection.

Phenomenon	Thermodynamic Explanation	Aerosol Explanation
“Warm rain” convection more prevalent over ocean	Reduced time for coalescence in stronger continental updraft	Larger droplets over ocean
Lightning activity more prevalent over Land	Greater instability over land and stronger cloud base updraft	Enhanced delivery of cloud water to mixed phase region
Large hail more prevalent over land	Stronger updraft and thinner warm rain region over land	Enhanced delivery of cloud water to mixed phase region
Upper tropospheric cirrus more prevalent over land	More vigorous ice production in stronger updraft over land	Greater numbers of small ice particles nucleated over land

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Table 3. Contrast in land-ocean physical and meteorological properties.

Property	Land	Ocean
Surface heat capacity	Small ($0.2\text{--}0.5\text{ cal gm}^{-1}\text{ }^{\circ}\text{C}^{-1}$), immobile surface	Large ($1\text{ cal gm}^{-1}\text{ }^{\circ}\text{C}^{-1}$), mobile surface
Air-surface temp. contrast	large ($1\text{--}10\text{ }^{\circ}\text{C}$)	Small ($\sim 1\text{ }^{\circ}\text{C}$)
Bowen ratio	Large (0.2–1)	Small (0.1)
Boundary layer CCN	High in polluted regions ($>1000\text{ cm}^{-3}$)	Low in clean regions ($\sim 100\text{ cm}^{-3}$)
Cloud base height	High (1000–4000 m)	Low (500 m)
Thickness of “warm” cloud	Small (0–3000 m)	Large ($\sim 4000\text{ m}$)
Cloud base updraft speed	High ($\sim 5\text{ m s}^{-1}$)	Small ($\sim 2\text{ m s}^{-1}$)
CAPE	Larger ($0\text{--}3000\text{ j kg}^{-1}$)	Smaller ($0\text{--}2000\text{ j kg}^{-1}$)
Convective updraft speed	High ($>10\text{ m s}^{-1}$)	Low ($<10\text{ m s}^{-1}$)

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The scientific basis for a satellite mission to retrieve CCN

D. Rosenfeld et al.

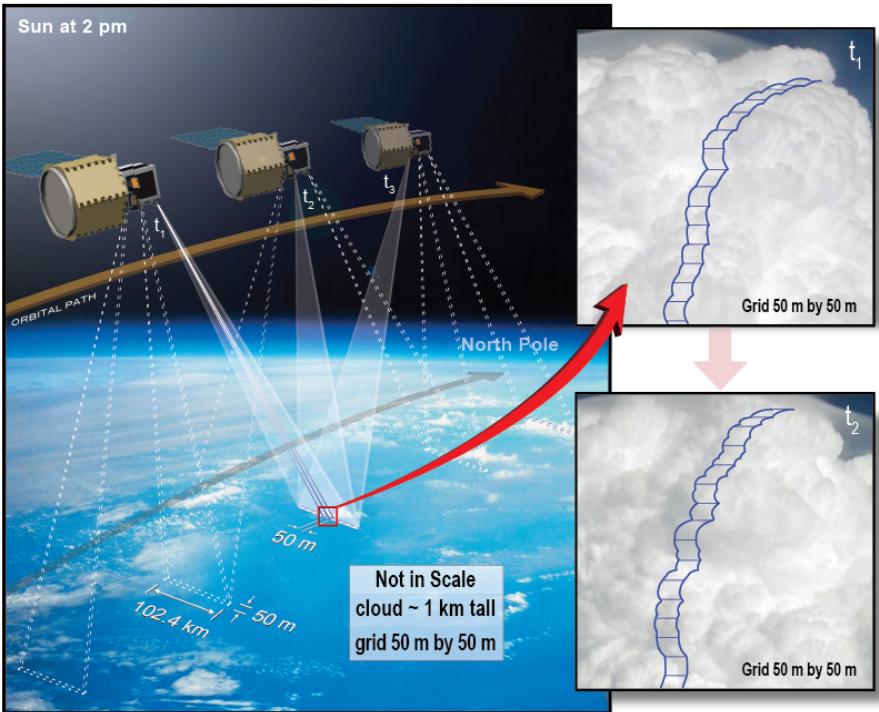


Fig. 1. An illustration of the measured vertical profiles of cloud parameters pointing 30° off track toward sun-illuminated cloud surfaces.

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Interactive Discussion

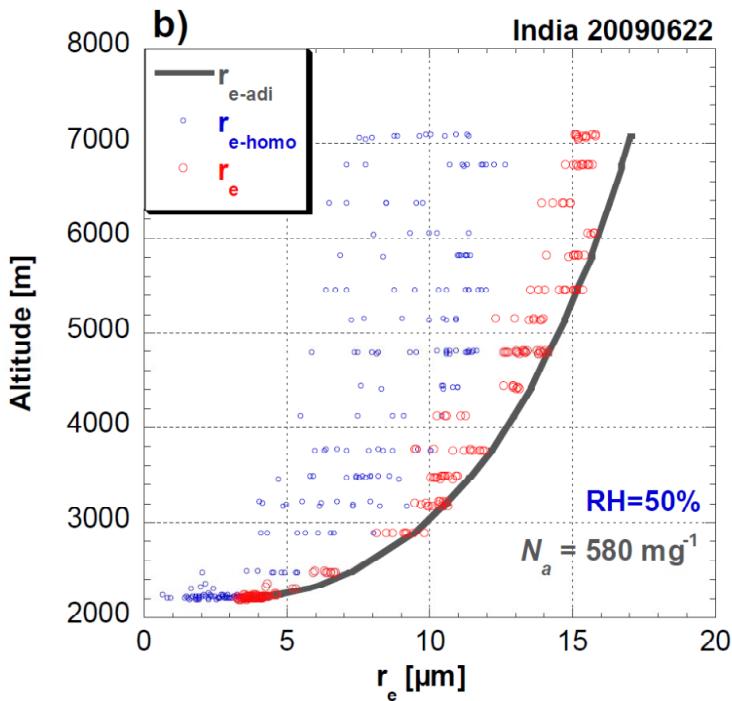


Fig. 2. Aircraft observations of tight relationships between r_e and altitude in a cluster of deep convective clouds over India. Each red circle marks the r_e measured in 1-s (~ 100 m) flight path. The dark curve shows the profile of the adiabatic r_e (r_{ea}). The N_a is calculated by the methodology described in Freud et al. (2011). It can be seen that for any given cloud depth the scatter of r_e is fairly small, and that values are normally within 2 μm from r_{ea} , despite including measurements with down to 5 % of adiabatic water. The blue circles indicate the expected effective radii in case of fully homogeneous mixing with entrained air which has 50 % RH, which was obtained from a nearby sounding. They are calculated for the mixing proportions of the actually measured adiabatic fractions. Similar relations are shown elsewhere in the world by Rosenfeld and Freud (2012).

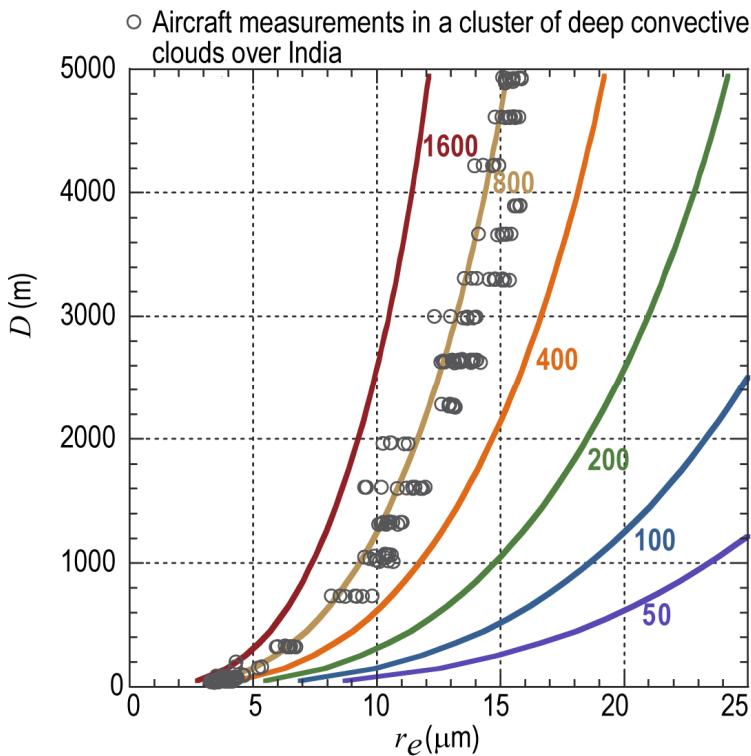


Fig. 3. Relationship between r_e and D for various values of N_a in cm^{-3} . The value derived from the profile of r_e measured by the aircraft is $N_a = 610 \text{ cm}^{-3}$ (580 mg^{-1}) at cloud base, within 5 % of the directly aircraft-measured maximum cloud base droplet concentration.

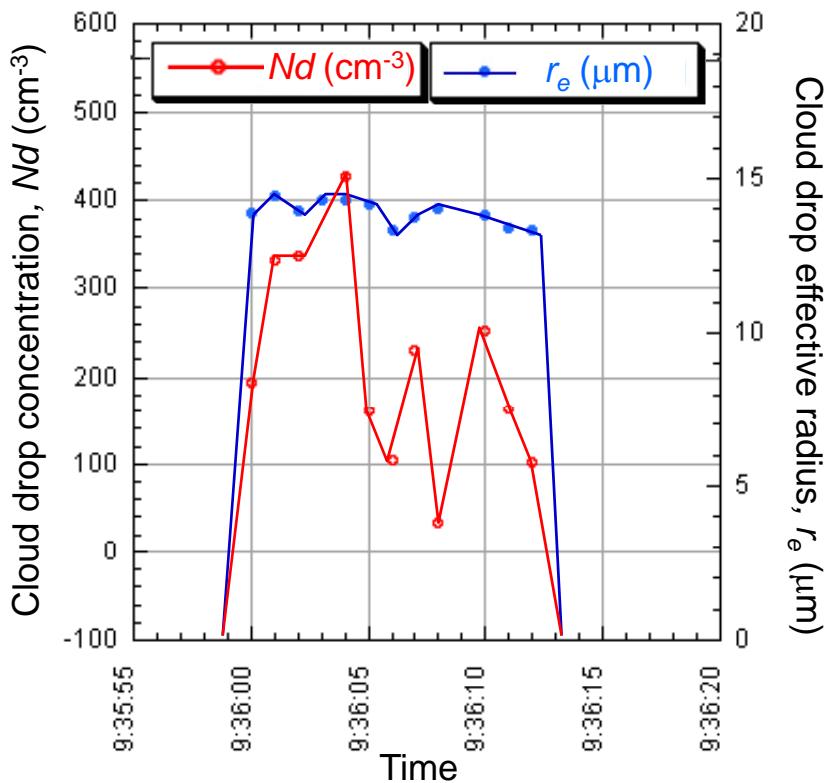


Fig. 4. The cloud drop concentration N_d and effective radius r_e in a horizontal cross section through a convective tower. Note the relative stability of r_e up to the cloud edge while N_d is highly variable. This is a manifestation of the nearly inhomogeneous mixing that dilutes N_d while keeping r_e nearly constant.

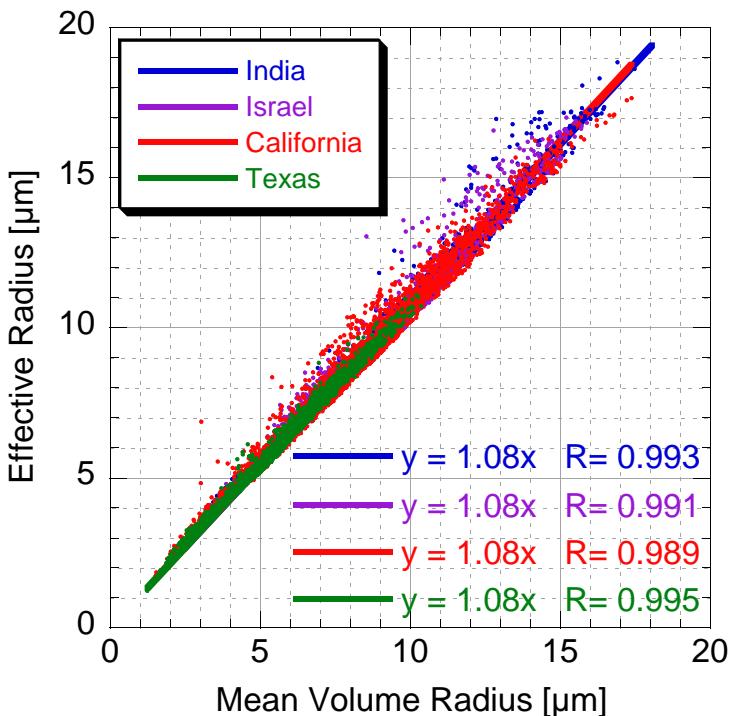


Fig. 5. The cloud droplet effective radius, r_e , versus the droplet mean volume radius, r_v , for 1-Hz averaged droplet size distributions measured in various locations, cloud types and by different cloud droplet probes. Note the universally highly repeatable and tight $r_e - r_v$ relationship (From Freud and Rosenfeld, 2011).

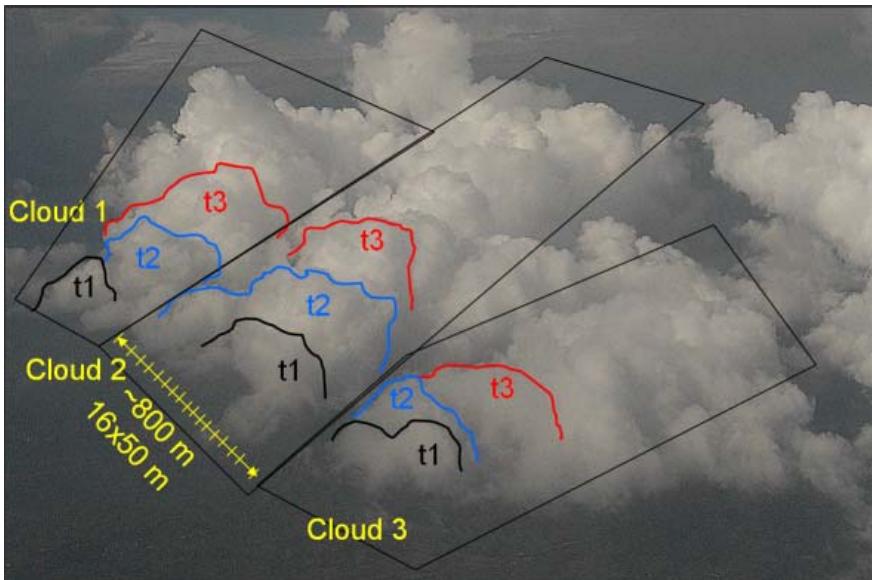


Fig. 6. Illustration of the topography of three small convective cloud segments. The contours show the possible development lines of the cloud top along three time steps, i.e. three views of the same cloud at t_1 , t_2 and t_3 . These identifiable cloud features just above cloud base develop vertically at a similar rate as the cloud base updraft. The MAI 50 m resolution is illustrated by the scale bar. The resolution appears to be sufficient for mapping the marked perturbations and track these features along a sequence of three 1-min images, and map the average topographies for t_1 and t_2 and for t_2 and t_3 . The differentiation of the two topographic maps will provide the cloud vertical motion. It appears that the retrieved updraft near cloud base, w_b , would be the largest for cloud 1 and the smallest for cloud 3.

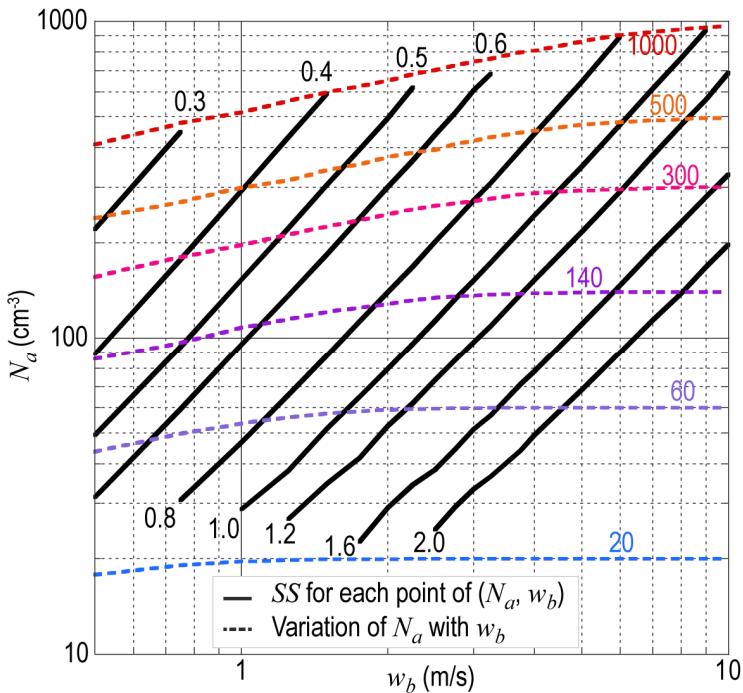


Fig. 7. The CCN spectra can be calculated using measurements or model simulations of N_a and $S(w_b)$. This chart was drawn using data from Pöschl et al. (2010) for the activation of organic aerosol particles with $\kappa = 0.13$ as observed for pristine tropical rainforest air. $\kappa = 0.3$ is more representative for most other continental aerosol regimes. Increasing κ would lower the S -values in the figure (Reutter et al., 2009).