

Estimation of the indirect radiative effects of aerosol on climate using a general circulation model



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Jesus College, Oxford
Hilary Term 2012

a thesis submitted for the degree of
Doctor of Philosophy



UNIVERSITY OF
OXFORD

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The indirect effects of anthropogenic aerosols—through their interactions with clouds—are currently one of the most uncertain perturbations to the radiative energy balance at the top of the atmosphere. A crucial link between aerosol and cloud is that aerosols can act as cloud condensation nuclei (CCN). This microphysical process must be parameterised if the large-scale effects are to be represented in a general circulation model (GCM). Theoretical work presented in this thesis highlights the importance of incorporating the kinetic limitations on droplet formation in aerosol activation parameterisations.

HadGEM-UKCA is a GCM, capable of representing the chemical and microphysical aerosol processes required to model CCN accurately. The author has incorporated a Köhler theory based parameterisation of aerosol activation into HadGEM-UKCA, to facilitate quantitative predictions of the indirect aerosol effects.

This thesis presents an estimate of the range of uncertainty in such predictions attributable to the choice of parameterisation of the sub-grid-scale variability of vertical velocity. Results of simulations demonstrate that the use of a characteristic updraught velocity cannot replicate results derived with a distribution of vertical velocities, and is to be discouraged in GCMs.

Consequently, work focuses on the effect of the variance (σ_w^2) of a Gaussian pdf of vertical velocity. Fixed values of σ_w and a configuration in which σ_w depends on turbulent kinetic energy are tested. Results from the mid-range fixed σ_w and TKE-based configurations both compare well with vertical velocity distributions and cloud droplet number concentrations measured *in situ*. However, the sparse set of available measurements does not provide enough of a constraint to recommend one or the other as the best configuration globally.

The radiative flux perturbation (RFP) due to the total effects of anthropogenic aerosol is estimated at -1.7 W m^{-2} for the TKE-based configuration. To the extent that it is valid to decouple the individual aerosol effects, the direct effect accounts for approximately -0.6 W m^{-2} of the total, the cloud albedo effect -0.8 W m^{-2} and the cloud lifetime effect -0.3 W m^{-2} , indicating that these effects are additive within HadGEM-UKCA. Total aerosol RFP ranges from -1.4 W m^{-2} from simulations with $\sigma_w = 0.1 \text{ m s}^{-1}$, up to -2.0 W m^{-2} for $\sigma_w = 0.7 \text{ m s}^{-1}$. This range of 0.6 W m^{-2} corresponds to almost a third of the total estimate of -1.9 W m^{-2} , obtained with the mid-range value of $\sigma_w = 0.4 \text{ m s}^{-1}$. Reducing the uncertainty in the parameterisation of σ_w is therefore an important step towards reducing the uncertainty in estimates of the indirect aerosol effects.

Acknowledgements

First and foremost I would like to acknowledge the support I have received from my supervisors. I would like to thank Philip Stier for his overall guidance of the project, his close supervision of my day-to-day work, and for his patience. I am very grateful to Don Grainger for his constructive input in the early stages, and his encouragement throughout. I would also like to say a special word of thanks to my supervisor at the Met Office, Andy Jones, for his thorough and rapid responses to queries, detailed and careful explanations, and valuable guidance, even from afar.

This work was funded by a studentship from the Natural Environment Research Council and a CASE award from the UK Met Office. I would therefore like to express my gratitude to NERC and the Met Office, but also to extend this gratitude to David Andrews, Stephen Blundell and Sarah Harrington, who were instrumental in securing this funding for me.

The CASE partnership has provided a unique insight into the Met Office and I am especially grateful to everyone in the Climate, Chemistry and Ecosystems group (now Earth Systems Science) who made me feel so welcome during my trips down to Exeter.

The work presented in this thesis would obviously not have been possible without the Unified Model and, despite all the grief its horrible user interface has caused me, I would of course like to acknowledge the work of literally hundreds of scientists in creating this model. More specifically, I would like to acknowledge the work of all of the members of the UKCA collaboration, in particular Colin Johnson and Graham Mann. I have received invaluable technical assistance from Mohit Dalvi, Luke Abraham and Paul Telford. I would like to thank Nicolas Bellouin for many useful discussions and advice, in person and via email. Outside of UKCA, I am also grateful to Adrian Lock for his assistance with coupling my scheme to the boundary layer turbulent kinetic energy.

Simulations on the supercomputer have only been possible due to the technical skills of the MONSooN team, and NCAS Computational Modelling Support, and I would like to acknowledge them here. In addition, I would like to express my sincere gratitude to Andy Heaps for his brilliant library of IDL routines, and the extremely clear guidebook which accompanies them. I am convinced that this library saved me hours and hours of work and I used it to produce nearly all of the figures in this document.

I am grateful for the interactions I have had with international collaborators. In particular, I had a thoroughly enjoyable dialogue with Donifan Barahona and Athanasios Nenes regarding the parameterisation development, and I have also had helpful advice from Steve Ghan.

Satellite datasets used in this thesis were organised by Benjamin Grandey, which has enabled the model evaluation in Chapter 3. Some of the data were processed by Johannes Quaas and Christopher Arnold, and I would like to thank them both. I want to thank Dominick Spracklen for providing me with his vast compendium of CCN measurements ahead of print, and would also like to acknowledge the efforts of all those involved in the flight campaigns discussed in Chapter 5.

It has been a remarkable privilege to work in the Atmospheric, Oceanic and Planetary

Physics sub-department over the course of this project, and I would like to thank everyone who has worked there during this time for making it such a pleasant, intellectually-stimulating, and memorable experience for me.

Within the department I would specifically like to thank all the past and present members of the Climate Processes Group for providing such a good working environment, for the countless informative discussions inside and outside of group meetings and for all kinds of other help that they have provided.

Within this group I have been especially fortunate to share my office with Benjamin Grandey and Zak Kipling to whom I am truly indebted. Benjamin's unfaltering enthusiasm for physics and his formidable work ethic have been great motivators for me, and he has taught me many things about IDL, aerosols, clouds, satellites and much more besides. I would also like to thank him for always being so thoughtful and cheerful in the office. More recently, Benjamin has even continued to provide assistance to me from his new home on the other side of the world!

Zak's computational powers are supreme, and I am particularly grateful to him for assimilating the UM into his brain during the first week of his DPhil, as this has made the last two and a half years of mine far, far less bewildering than the pre-Zak era. Not only that, but Zak must have the patience of a saint, because he has never once complained about my interruptions and always seems happy to provide kind and patient explanations to my queries. Finally, I would like to thank him for his willingness to enter into discussions about physics, maths, meteorology, current affairs, music, bicycles and numerous other distractions.

Throughout my time in Oxford, Stephen and Katherine Blundell have been greatly encouraging to me, and have provided me with two exceptional role models, and I would like to acknowledge that here.

Science can be a tedious and painstaking business, and I would like to thank all the members of AOPP who have provided much-needed non-scientific relief over tea and biscuits. Likewise, I would like to thank the condensed matter physicists for allowing me to join their lunch-breaks in the Clarendon.

Throughout my life, music has provided me with a wealth of friends, enjoyment and distraction from work, and on that note I would particularly like to thank Theo Lester, W. A. Mozart and the cast of *The Marriage of Figaro*, who provided such welcome music, dancing and fireworks during a rather cold and dark winter.

I am truly grateful for the support I have received from my friends, and I would especially like to thank Tom Fuller and Miranda Laurence for their steadfast friendship over the past few years. As well as frequently taking Andrew off my hands, Tom has also proof-read part of this thesis, and Miranda has provided much-needed encouragement throughout.

I am deeply grateful to my parents, Janet and Gordon West, my brothers Adrian and Alexander, and Grandma Kay for their enduring love and support.

Last of all, I must acknowledge Andrew Steele for the domestic, technical and emotional support he has provided throughout my DPhil, and especially while I have been writing up. More generally, I want to thank Andrew for sharing with me some of his enthusiasm for science, and reminding me of my own; for going halves with me on a bicycle made for two, and for the adventures that followed; and most of all for his unwavering belief that I could do this work and write about it.

This thesis is dedicated to my grandfather, Professor Eric A. Lunzer, whose passion for learning, approach to life, and intellectual endeavours across a range of disciplines remain a constant source of inspiration to me.



Henry Purcell, *Dido and Aeneas*
Act II, scene ii

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Introduction

Quantifying the effects of aerosol on the Earth's climate remains one of the largest challenges in contemporary climate science. One way to characterise our changing climate is to consider the energy balance between incoming and outgoing radiation, and aerosols may affect this energy balance both directly, by scattering and absorbing radiation, and indirectly, through their interactions with clouds. About two-thirds of the Earth's surface is covered by cloud at any one time, and therefore understanding how this cloud interacts with radiation is vital.

A crucial link between aerosols and clouds is the ability of aerosols to act as condensation nuclei, on which cloud droplets form. This process is manifestly microphysical (with aerosols, cloud droplets and raindrops ranging in size from nanometres to millimetres), whilst the cloud systems they comprise can extend tens of kilometres up into the atmosphere and cover hundreds of square kilometres horizontally. This collision of scales poses significant difficulties for both observing and modelling these aerosol–cloud interactions. As a consequence, the uncertainty in current estimates of the indirect radiative effects of aerosol is the largest of all known factors affecting the Earth's radiative balance. (The Fourth Assessment Report of the Intergovernmental Panel on Climate Change quoted an error bar on indirect aerosol effects which was comparable to the total radiative effect of carbon dioxide.)

Reducing this uncertainty is therefore of paramount importance. The work in this thesis attempts to tackle this problem by improving the representation of indirect aerosol effects in a computational climate model. One of the difficulties in modelling aerosol effects on clouds is that these microphysical processes are impossible to explicitly include in a global climate model, which is typically evaluated on a model grid with a horizontal resolution of tens to hundreds of kilometres. As such, various parameterisations have been developed which attempt to enable the sub-grid-scale process of cloud droplet formation to be represented in large-scale global models. These parameterisations can be evaluated against a variety of empirical observations ranging in scale from lab-based process studies to global satellite datasets.

This thesis is divided into the following chapters: the remainder of **Chapter 1** provides an introduction to aerosols and an overview of the relevant aspects of warm cloud formation, including Köhler theory, and aerosol activation parameterisations; **Chapter 2** focuses on the intercomparison of two of these parameterisations and the development of one of them to properly account for the effects of inertially-limited droplets; **Chapter 3** describes how one of these parameterisations of cloud droplet formation has been implemented into a general circulation model (GCM) in order to make estimates of the indirect radiative

effects of aerosol. This chapter also includes an evaluation of modelled cloud condensation nuclei against surface measurements and of several modelled cloud and aerosol properties against similar quantities retrieved from three Earth-observing satellite datasets; **Chapter 4** discusses the representation of sub-grid-scale variations of vertical velocity within a GCM, and defines five different configurations to be tested; **Chapter 5** evaluates the behaviour of each of these model configurations against *in situ* observations from a dozen aircraft flight campaigns; **Chapter 6** quantifies the radiative effects of anthropogenic aerosol in each model configuration, discusses their similarities and differences, and estimates the uncertainty introduced by different parameterisations of updraught velocity. It also places this estimate into the context of ranges of aerosol effects made by other recent intercomparison studies; finally, **Chapter 7** summarises the work contained in the preceding chapters, draws together the conclusions from each of these, and makes some suggestions for the future direction of work in this field.

All GCM simulations in this work are carried out with HadGEM3-A, the UK Met Office Hadley Centre Global Environmental Model in atmosphere-only mode (Hewitt et al., 2011), coupled to the new UK Chemistry & Aerosols community model, UKCA, which has been undergoing ongoing developments throughout the course of this project in a collaboration between scientists from the Met Office and the Universities of Leeds, Cambridge and Oxford. The active contribution of the author to this collaboration is in the implementation of an aerosol activation scheme which links the aerosol modelled by UKCA to the radiation and large-scale precipitation schemes in HadGEM3, thus allowing the indirect aerosol effects to be modelled. This aspect of the model is referred to as UKCA-Activate.

1.1 Atmospheric aerosols

Aerosols are liquid or solid particles suspended in the air. This suspension of particles plays an important role in atmospheric chemistry, and also has implications for human health, due to its impact on air quality. However, the rest of this work is principally concerned with the interaction of aerosols with radiation, which can be divided into direct and indirect effects, described below in Sec. 1.2.1 and Sec. 1.2.2. While some aerosols are natural in origin, and occur in the atmosphere due to a variety of natural processes such as the eruption of volcanoes, sea spray and mineral dust lifted by the wind, the global mean aerosol burden of the atmosphere has increased considerably due to industrialisation (Lohmann et al., 2007). The combustion of fuel causes a significant amount of anthropogenic aerosol to be added to the concoction, and anthropogenic aerosols now play a significant role in the Earth's radiative balance.

1.1.1 Aerosol composition

Atmospheric aerosols can be categorised by their origin into primary and secondary aerosols. Primary aerosols are those which are directly emitted as particulates, through the disintegration and dispersion of solids and liquids, whereas secondary aerosols are formed through the condensation of compounds from the gas phase (Andreae and Rosenfeld, 2008). Once airborne, aerosol particles may undergo a series of changes as their size and composition are modified by their interactions with the surrounding environment and one-another. During their short atmospheric lifetime of a few days to a few weeks, possible interactions include: condensation of gases onto existing particles, evaporation, chemical reactions, coagulation with other particles or activation to become cloud droplets. As a result of these processes, most aerosols are composites of various chemical components,

both soluble and insoluble. Aerosols are finally removed from the atmosphere due to their incorporation into cloud droplets and subsequent precipitation (wet deposition), or by sedimentation (dry deposition) (Seinfeld and Pandis, 1998).

Aerosols are traditionally categorised into fine particles (diameter, $D < 2.5 \mu\text{m}$) and coarse particles ($D > 2.5 \mu\text{m}$). The fine particles are further split into the Aitken mode ($0.005 < D < 0.1 \mu\text{m}$) and the accumulation mode ($0.1 < D < 2.5 \mu\text{m}$). While the Aitken mode accounts for the largest number of particles, it only accounts for a few percent of the mass. The accumulation mode usually contains the greatest fraction of surface area, and a significant proportion of the mass of the distribution. The bulk of the mass is usually contained within the coarse mode. The Aitken mode is typically partitioned further, especially in models, such that the large number of very small particles that nucleate directly from the gas phase can be considered in their own ‘nucleation’ mode, giving four size-modes in total. However, since the fate of the tiny and numerous nucleation particles is predominantly to coalesce to form bigger particles and hence transfer to the Aitken mode, three modes is also a common representation.

Since the geographical variation in aerosol sources is so great, and the lifetime of aerosols in the troposphere relatively short, the concentration and composition of aerosols in the troposphere varies considerably depending on location and time. In terms of modelling, a detailed emissions dataset is a necessary but not sufficient prerequisite for the practical incorporation of the aerosol activation process into a global model. What is needed additionally is knowledge of the spatial and temporal evolution of the distribution of potential cloud condensation nuclei (CCN), in the form of a comprehensive, interactive aerosol–chemistry transport model (Andreae and Rosenfeld, 2008).

1.1.2 Aerosol size distributions

Aerosol particles cover a vast size range, from a few nanometres up to tens of microns for large particles (such as those emitted directly from combustion, sea salt or mineral dust). In the troposphere, the density of aerosol particles varies from about 1 cm^{-3} in a clean marine environment such as the southern oceans to over $10\,000 \text{ cm}^{-3}$ in heavily-polluted urban regions such as Guangzhou and Beijing [Andreae and Rosenfeld 2008]. Of course, it would be computationally extortionate to attempt to track each particle individually, so instead various attempts have been made to mathematically describe aerosols with a number–size distribution.

In a sectional representation, aerosol size range is divided into discrete intervals, with the number concentration in each interval, N_i , normalised by the width of the interval, Δa , such that the value of the aerosol distribution for a given interval, i , is $n_i = N_i / \Delta a$. In this case the area beneath the curve is proportional to the number concentration. As the size intervals tend to the infinitesimal limit, $\Delta a \rightarrow da$, the size distribution can be defined as $n(a)da$, the number of particles per unit volume in the size interval a to $a + da$. Hence, the total number concentration of particles is

$$N = \int_0^{\infty} n(a)da, \quad (1.1)$$

assuming the number distribution is a continuous function of radius (valid beyond a certain number of particles (Seinfeld and Pandis, 1998)). Equivalently,

$$n(a) = \frac{dN}{da}. \quad (1.2)$$

Since the aerosol size distribution generally covers many orders of magnitude, a logarithmic scale is often convenient, where the distribution function $n_e d(\ln a)$ is the number concentrations of particles with radius in the interval $d(\ln a)$ such that the cumulative distribution and total number concentration are

$$n_e(a) = \frac{dN(a)}{d(\ln a)} \quad \text{and} \quad N = \int_{-\infty}^{\infty} n_e(\ln a) d \ln a. \quad (1.3)$$

These distributions describe how the number concentration of particles varies as a function of radius (or log-radius). In the early stages of aerosol science, aerosol number-size distributions were often plotted as $\frac{dN(a)}{d(\ln a)}$ against $\ln a$. As shown in Fig. 1.1(a), this results in a near straight line, and led to the representation of size distributions by a power law or Junge distribution (Junge, 1953; Clark and Whitby, 1967), where

$$\frac{dN(a)}{d(\ln a)} = Ca^{-\alpha} \quad (1.4)$$

and the slope of the line on a log-log plot is given by $-\alpha$. However, assuming the initial dry aerosol particles are spherical, size distribution in terms of number is trivially transformable to a corresponding size distribution in terms of surface area or volume. Once this transformation is performed, and the aerosol distribution replotted in terms of surface area or volume, as shown in Fig. 1.1(d) and (e), it becomes apparent that those minor deviations from the straight line of the power law are actually significant modes. Amongst others, Whitby proposed that these modes can be represented by a sum of three log-normal distributions (Whitby, 1978).

In a log-normal distribution $f(x)$, the logarithm of x is normally distributed, such that if $f(x)$ is plotted against $\ln x$, the familiar bell-shaped curve is retrieved (Aitchison and Brown, 1957). The distribution can be defined simply by the geometric mean value of x , and the geometric standard deviation of the distribution, σ_g . Atmospheric aerosol size distributions are often described by the sum of I log-normal modes,

$$\frac{dN(a)}{d(\ln a)} = \sum_{i=1}^I \frac{N_i}{\sqrt{2\pi \ln \sigma_i}} \exp \left\{ -\frac{\ln^2 (a/a_{m,i})}{2 \ln^2 \sigma_{g,i}} \right\}, \quad (1.5)$$

where N_i , $a_{m,i}$ and $\sigma_{g,i}$ are the total number concentration, geometric mean dry radius¹ and geometric standard deviation of aerosol mode i , respectively (Von der Emde and Wacker, 1993; Abdul-Razzak and Ghan, 2000). A trivial transformation converts this log-normal number distribution to a surface or volume distribution. One of the great advantages of this function is that such moment distributions (i.e. related by a power law) of the original log-normal distribution are also log-normal, and share the same geometric standard deviation (Hatch and Choate, 1929; Seinfeld and Pandis, 1998; Hinds, 1999).

1.2 Aerosol radiative effects

In order to quantify and compare the different radiative effects of atmospheric aerosol with one-another and with other drivers of change within the climate system, a quantitative metric is needed. The concept of radiative forcing is explored below, although, as detailed in Chapter 6, if all of the indirect aerosol effects are to be included within the forcing estimate,

¹Note that the geometric mean is identical to the median, but not the mode, for a log-normal distribution.

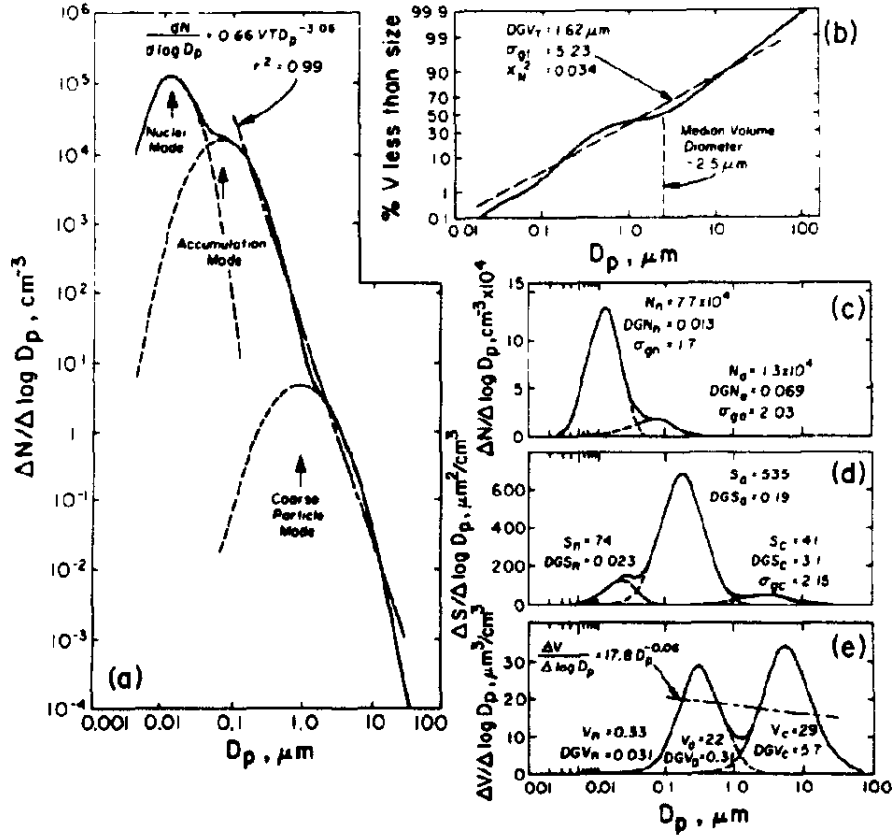


Figure 1.1: Average urban aerosol size distributions plotted in five different ways. In (a), a power law function has been fitted to the number distribution over the size range $0.0 < D < 32.0 \mu\text{m}$. (b) shows the percentage difference between volume and diameter as a function of diameter. In (c) a log-normal distribution has been fitted to the range $0.1 < D < 32.0 \mu\text{m}$. (d) shows the transformation of this distribution to surface area and (e) the transformation to volume. It is evident that the modal nature of the aerosol is shown best by the log-normal representation of (c), (d) and (e). From Whitby (1978).

a modified version of this concept, known as a radiative flux perturbation, is necessary and must be calculated instead.

Radiative forcing

The climate of the Earth is a complex system of myriad interacting processes and feedback loops. In order to discern and quantify the influence of a particular one of these processes on the radiative energy balance of the Earth it is necessary to define a standard property which may be compared between factors. Forster et al. (2007) define radiative forcing as 'the change in net (down minus up) irradiance (solar plus longwave in W m^{-2}) at the tropopause after allowing for stratospheric temperatures to readjust to radiative equilibrium, but with surface and tropospheric temperatures and states held fixed at the unperturbed levels'. In other words, the radiative forcing due to a particular process is the change in the Earth's energy balance that would occur if that factor alone were altered, and all other variables were held constant. A positive radiative forcing indicates an increase in the energy of the system leading to a warming effect, while negative indicates cooling. Radiative forcing is

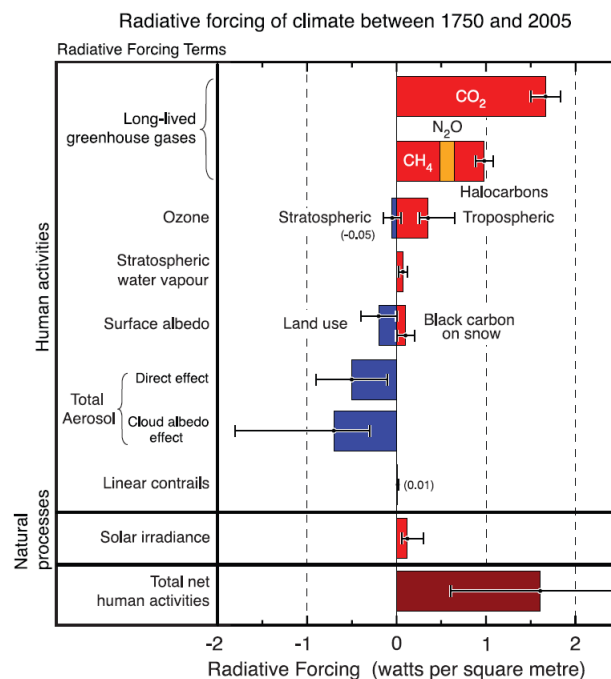


Figure 1.2: Summary of the principal components of the radiative forcing of climate change. The values represent the forcings in 2005 relative to 1750. The current range of uncertainty in the radiative forcing of each component is shown by a black error bar. From Forster et al. (2007).

often used to assess the impact different factors have on climate, by comparing the total radiative forcing relative to the beginning of the industrial revolution (starting in about 1750).

Fig. 1.2 shows the global mean total radiative forcings over the period 1750 to 2005 for various climate forcing agents and mechanisms. Currently, the uncertainty in the radiative forcing due to the indirect aerosol effect is the greatest uncertainty amongst the known causes of radiative forcing. Reducing this level of uncertainty will reduce the overall uncertainty in radiative forcing of the climate and provide tighter constraints for climate modelling and prediction.

1.2.1 Direct effects

The direct effect of aerosol particles on the radiative balance of the Earth involves a combination of scattering and absorption of radiation by the aerosol particles themselves. As shown in Fig. 1.2, this effect is estimated to contribute a net radiative forcing of -0.5 W m^{-2} with a 5 to 95% confidence range of -0.1 to -0.9 W m^{-2} , a net cooling effect (Forster et al., 2007). The 'semi-direct effect' is the mechanism by which absorption of shortwave (solar) radiation by tropospheric aerosols leads to heating of the troposphere that in turn changes the relative humidity and the stability of the troposphere (Forster et al., 2007). This could hinder cloud formation, and/or cause existing cloud droplets to evaporate (Lohmann et al., 2007). The semi-direct effect is not strictly classed as a radiative forcing by the IPCC (Forster et al., 2007), since it does not leave the atmospheric state unperturbed, due to modifications to the hydrological cycle, which is why it does not feature in Fig. 1.2.

1.2.2 Indirect effects

Aerosols also affect the radiative balance due to their interactions with clouds. The presence, lifetime, abundance and properties of clouds may all be modified by the aerosols present in the surrounding environment. In the Earth's atmosphere, warm cloud formation itself is dependent on the presence of aerosols to act as condensation nuclei for droplet formation, and this process will be considered in more detail in Sec. 1.3.2. Aerosols can also act as nuclei for ice crystal formation, and indirect aerosol radiative effects can thereby occur due to the effects of anthropogenic aerosols on the radiative properties of mixed-phase and ice clouds, but these effects are beyond the scope of the work in this thesis, which is restricted to the effects on warm clouds.

Indirect aerosol effects on warm clouds occur when increasing the number of cloud condensation nuclei leads to an increase in the number of cloud droplets. In a cloud of constant liquid water content, the mean size of the droplets is thereby reduced. This reduction in droplet size has two main effects.

First, it leads to an increase in the total surface area of the droplets (in a cloud of constant liquid water content), such that the cloud reflects more of the incoming solar radiation back to space, leading to a net cooling. This effect is known as the **cloud albedo effect** (but may also be referred to as 'the Twomey effect' or simply 'the first indirect effect' in the literature) (Twomey, 1959, 1974; Warner, 1968).

Second, a decrease in the mean droplet size will decrease the efficiency with which precipitation forms in a cloud, which may cause the cloud to persist for longer, and is hence known as the **cloud lifetime effect** (Albrecht, 1989). Albrecht proposed that increasing the number of CCN in marine stratiform clouds would suppress the rate of drizzle formation, thus reducing the rate at which water was dissipated from the cloud and hence increasing both cloud liquid water content and cloud lifetime. This would lead to a net cooling effect due to the increased persistence of reflective clouds².

Several measurement studies have made observations consistent with this mechanism in marine stratiform clouds, in particular, evidence of reduced droplet sizes, suppression of drizzle and increased liquid water path (column-integrated liquid water content) in the vicinity of enhanced aerosol concentrations in ship tracks has been found in a number of studies [e.g. Ferek et al. 1998, 2000; Peng et al. 2002]. Additionally, an *in situ* flight campaign over the Amazon also found evidence that heavy smoke from forest fires can lead to a decrease in cloud droplet size and a suppression of precipitation (Andreae et al., 2004).

However, other studies have found that polluted clouds can have less liquid water than clean clouds [e.g. Platnick et al. 2000; Coakley and Walsh 2002], although this can be attributed to the enhanced entrainment of dry air into polluted clouds, leading to subsequent evaporation of cloud droplets (Ackerman et al., 2004). Increasing the cloud lifetime may also increase the height to which a cloud grows, potentially by invigorating deep convection, although the mechanism by which this occurs is contested in the literature (Pincus and Baker, 1994; Andreae et al., 2004; Koren et al., 2005, 2010; Rosenfeld et al., 2008). In short, there is conflicting evidence of the impacts of changes to cloud precipitation efficiency due to aerosols, which can be summarised by concluding that the response of a cloud to changes in the droplet number is sensitive to the local meteorological conditions (Ackerman et al., 2004).

Nevertheless, the physical process that needs to be encoded explicitly in a model is

²An increase in the liquid water content would cause a cloud to reflect more incoming shortwave radiation (hence the cloud appears brighter), leading to a cooling effect, although increasing the liquid water content could also lead to the cloud exerting a longwave warming effect (Jones et al., 2001).

the effect that droplet size has on precipitation efficiency. When the atmosphere is in equilibrium, the global annual mean precipitation rate balances the evaporation rate at the surface. In order for these two processes to remain in equilibrium when the precipitation efficiency changes (due to a change to the number and size of cloud droplets), the liquid water content of the cloud must respond accordingly to counteract this change and maintain the balance.

As discussed by Jones et al. (2001), the formation of precipitation in a non-freezing cloud depends upon the runaway growth of a small fraction of cloud droplets that grow to such a size that they fall faster, relative to other smaller droplets. They collide with these smaller droplets, and coalesce, eventually growing large enough to fall out as precipitation. This process is referred to as the **autoconversion of cloud water to rain water**, and is slower for a higher concentration of small droplets than for a lower concentration of large droplets (for a given total volume of water). The reason for this is two-fold: first, smaller droplets are less prone to collisions, because they tend to flow around one-another rather than collide and coalesce; second, more collisions are required of small droplets in order for them to grow to raindrop size. Thus, if we compare the precipitation efficiency (i.e. the rate of precipitation per unit of liquid water), it will be lower for the case of more, smaller droplets than for the case of fewer, larger droplets.

As a result, in a world with more, smaller cloud droplets, the liquid water content of clouds must increase in order to maintain the balance of the global hydrological cycle. In practice, the global evaporation rate remains roughly constant, and therefore the overall precipitation rate must also remain constant to retain the balance. It then follows that the cloud liquid water content must be higher in a world with more, smaller droplets than one with fewer, larger droplets, to compensate for the lower precipitation efficiency in the former. This is the physical basis of the cloud lifetime effect.

Due to the difficulty in parameterising the microphysical processes on which both indirect aerosol effects depend, and uncertainties in the inputs to these parameterisations, modelling estimates of the indirect aerosol effects are highly uncertain. Constraining the estimate of the overall effects from inverse calculations from present-day satellite studies is limited by the difficulty of performing satellite retrievals of aerosol–cloud interactions (Quaas et al., 2006). Nevertheless, the IPCC reports that clear progress was made between the Third and Fourth Assessment Reports, and provides a best estimate of the net radiative forcing due to the cloud albedo effect from models to be -0.7 W m^{-2} with a 5 to 95% confidence range of -0.3 to -1.8 W m^{-2} , as shown in Fig. 1.2 (Forster et al., 2007).

At the time of the Third Assessment Report, most models were only capable of simulating the indirect radiative effects of sulphate aerosols, whereas by the time of the Fourth Assessment Report, most models simulated external mixtures of multiple aerosol species, incorporating various combinations of sulphate, sea salt, black carbon, organic carbon and dust. A minority of models also included nitrates and some models had the facility of dynamically-evolving external mixtures of internally-mixed lognormal modes.

A second major area of progress between the two reports was in the modelling of aerosol activation itself. Most of the models in the Third Assessment Report relied upon empirical relationships between the mass (or number) of aerosols and the cloud droplet number concentration, with a few exceptions. However, a significant fraction of models included in the Fourth Assessment Report had incorporated physically-based parameterisations of aerosol activation. The evolution of the representation of aerosol activation from empirical methods through to the continued development of modern parameterisations is discussed in more detail in Sec. 1.4 and Chapter 2.

Incorporating the influence of cloud droplet number concentration and liquid water

content on the autoconversion rate in a climate model allows the cloud lifetime effect to be simulated. However, the cloud lifetime effect cannot be quantified as a strict radiative forcing, owing to its implicit modification to the hydrological cycle. Instead, alternative metrics have been proposed to allow the radiative effects of this mechanism, and others involving feedbacks, to be quantified. Current thinking has converged upon the Radiative Flux Perturbation (RFP) as the most suitable of these metrics for estimating the total radiative effects of anthropogenic aerosols since pre-industrial times (Lohmann et al., 2010). The RFP is obtained by taking the difference in annual mean net radiation at the top of the atmosphere between a simulation with present-day aerosol emissions, and one with pre-industrial aerosol emissions. Estimates of radiative flux perturbations due to anthropogenic aerosol will be returned to in more depth in Chapter 6.

1.3 Warm cloud microphysics

The ability of a GCM to model the indirect effects of aerosols on warm clouds partly depends on the manner in which the process of aerosol activation is parameterised. In order to make sense of the parameterisations of aerosol activation for a GCM, it is important to understand the physical processes governing cloud droplet formation. A brief review of the Köhler theory of hygroscopic droplet growth follows, as this is the foundation of all aerosol activation parameterisations. Full details are available in Köhler (1936), and more recent developments in dynamic Köhler theory can be found in Pruppacher and Klett (1997).

1.3.1 Warm cloud formation

The formation of clouds in the atmosphere is possible due to a combination of thermodynamic and microphysical processes. For a cloud to form, it is necessary for a large volume of moist air to be cooled below its dew point. The dew point temperature, T_d , is defined as the temperature to which moist air must be cooled, at constant pressure and mixing ratio, for it to reach saturation with respect to water. (Where mixing ratio, w , is the ratio of mass of water vapour, M_v to mass of dry air, M_d , per unit volume of moist air, $w = M_v/M_d = \rho_v/\rho_d$.) A parcel of air which is saturated with respect to water defines the concept of relative humidity, since relative humidity, f , is defined as the mixing ratio of a parcel at a particular temperature and pressure, compared to its saturation mixing ratio value at the same temperature and pressure,

$$f = \frac{w}{w_s(p, T)} \approx \frac{e}{e_s} \quad (1.6)$$

where e is the vapour pressure of water in the parcel, and e_s is the saturated (equilibrium) vapour pressure of water over a plane surface of water at the temperature of the parcel. A parcel is described as supersaturated when $e > e_s$. The temperature dependence of e_s is defined by the Clausius–Clapeyron equation:

$$\frac{de_s}{dT} = \frac{L}{T(v_v - v_l)}, \quad (1.7)$$

where L is the latent heat of vaporisation, and v_v and v_l are the specific volumes of water vapour and liquid water. In ordinary atmospheric conditions, $v_v \gg v_l$ and water behaves as an ideal gas, ($e = \rho_v R_v T$, where R_v is the ideal gas constant), so Eq. (1.7) reduces to

$$\frac{de_s}{dT} = \frac{L}{T v_v} = \frac{L e_s}{R_v T^2}. \quad (1.8)$$

The temperature dependence of L makes an analytic expression for $e_s(T)$ impossible, but various approximations can be made depending on the applicable temperature range (Goff and Gratch, 1946; Rogers and Yau, 1989).

In the atmosphere, the most prevalent mechanism for a parcel of moist air to be cooled below T_d is via the approximately adiabatic expansion of ascending air, although it can be caused by radiative cooling or due to the convergence of air parcels with different temperatures and moisture contents. In an ascending parcel, the updraught velocity affects the rate of cooling and hence the amount of condensation which forms. However, the condensation of water into individual cloud droplets is governed by complex processes on a far smaller scale (Rogers and Yau, 1989).

1.3.2 Cloud droplet formation

In supersaturated air, cloud droplets may form by either of two microphysical processes. Homogeneous nucleation is the nucleation of water vapour onto embryonic droplets themselves consisting only of water (Seinfeld and Pandis, 1998). This requires the initial formation of tiny embryonic droplets (clusters of water molecules) with a very tightly curved surface. The effect of this curvature is that the saturated vapour pressure over the embryo is much higher than that over a plane surface of water, as described by the Kelvin effect (Sec. 1.3.3). As a result, the saturation ratio (actual vapour pressure divided by saturated vapour pressure over a plane surface) required for droplets to form on these clusters is predicted to be of the order 3.5 to 8. Experimentally, relative humidities of over 800% have been reached, in air free of all impurities and ions, before cloud droplets will form spontaneously in an expansion chamber (Pruppacher and Klett, 1997).

The abundance of aerosols in the atmosphere provides a mechanism for droplet formation at far lower supersaturation. As a result, levels of supersaturation required for homogeneous nucleation are never reached (Andreae and Rosenfeld, 2008). Heterogeneous nucleation, the condensation of water on a foreign substance or surface, is favourable because of the reduction in saturated vapour pressure over the droplet either due to the presence of a solute or due to the opposition of the Kelvin effect provided by the presence of an insoluble core which increases the radius of curvature of the droplet.

1.3.3 Equilibrium Köhler theory

Köhler theory of droplet formation describes how the rate of water condensation onto a particle in equilibrium with its surroundings is dependent on both the solubility and radius of curvature of that particle. Thus the formation of cloud droplets is also dependent on the chemical composition and size distribution of the ambient aerosol, and an aerosol activation parameterisation should seek to incorporate this dependence, if it is to be physically realistic.

Curvature effect

For droplets to grow, the rate of condensation of water onto the droplet must exceed the evaporation from the surface. First, we consider the process of homogeneous nucleation on a spherical droplet of water. The vapour pressure over a curved surface as a function of radius, $e_s(r)$ will be discussed with respect to the vapour pressure over a plane surface, $e_s(\infty)$. Defining the saturation ratio as $s = e/e_s(\infty)$, the energy associated with the homogeneous nucleation of a spherical droplet can be written as

$$\Delta G = -\frac{4\pi r^3}{3v_l}kT \ln s + 4\pi r^2\sigma, \quad (1.9)$$

where k is the Boltzmann constant, T is temperature, v_l is the volume occupied by a molecule in the liquid phase and σ is the surface tension. (The derivation of Eq. (1.9) from the thermodynamic relation for the Gibbs free energy is given in Appendix A.)

The sign of the first term on the right hand side of Eq. (1.9) is defined by the saturation ratio, and this has a strong effect on the behaviour of ΔG as a function of r . In sub-saturated conditions, $e < e_s(\infty)$ so $\ln s$ is negative, and hence ΔG is always positive and increases with increasing r . This inhibits droplet formation under subsaturated conditions. In supersaturated conditions, $e_s > e_s(\infty)$ so $\ln s$ is positive. In this case, ΔG can be positive or negative, depending on the value of r . Initially, ΔG increases with increasing r , inhibiting the growth of very small particles in supersaturated conditions. ΔG reaches a maximum at $r = r_c$ and then decreases with further increase in r , allowing a droplet to continue growing spontaneously through condensation from the vapour phase, since further growth causes a further drop in ΔG . The maximum is a metastable equilibrium, and its location can be found by setting $\partial\Delta G/\partial r = 0$ in Eq. (1.9) to obtain

$$r_c = \frac{2\sigma v_l}{kT \ln s}. \quad (1.10)$$

This equation relates the equilibrium radius of a droplet of pure substance (in this case, water) to the physical properties of that substance (the surface tension, σ , and the volume of a molecule in the liquid phase, v_l) and the saturation ratio over the droplet in its environment. It can be re-expressed in terms of molar units as the more familiar Kelvin equation

$$\frac{e_s(r)}{e_s(\infty)} = \exp\left(\frac{2\sigma M_w}{\rho_w R T r}\right) = \exp\left(\frac{A}{r}\right), \quad (1.11)$$

where M_w is the molar mass of water, ρ_w is the density of water and

$$A = \frac{2\sigma M_w}{\rho_w R T}. \quad (1.12)$$

It is apparent from the Kelvin equation that the vapour pressure over a curved surface of a substance is always greater than the vapour pressure over a plane surface of the same substance (Seinfeld and Pandis, 1998; Wallace and Hobbs, 2006). Ultimately, because of this Kelvin effect, the larger a droplet, the faster it will grow (by volume). This will be discussed further below.

Solute effect

As it happens, many atmospheric aerosols are soluble in water, and form a droplet of aqueous solution as water condenses on to them. In terms of droplet growth, this solubility is fortunate because the vapour pressure over a solution is lowered by the presence of the solute and hence growth is energetically favourable. The fractional reduction in vapour pressure due to the presence of a solute is given by Raoult's law, which states that the vapour pressure of an ideal solution is dependent on the vapour pressure of each chemical component and the mole fraction of the component present in the solution, so

$$\frac{e'}{e_s(\infty)} = \frac{n_w}{n_w + n_s}, \quad (1.13)$$

where e' is the vapour pressure of a solution consisting of n_w moles of water and n_s moles of dissolved solute. For a dilute solution, where $n_s \ll n_w$, we may expand binomially to give

$$\frac{e'}{e_s(\infty)} \approx 1 - \frac{n_s}{n_w}. \quad (1.14)$$

where the number of moles of solute, $n_s = \nu m_s / M_s$, where m_s is the mass of solute, M_s is the molar mass of solute and ν represents the degree of ionic dissociation of the solute and moles of water is given by $n_w = m_w / M_w$. The mass of solute present is given by $m_s = \frac{4}{3}\pi a^3 \rho_s$ where a is the radius of the dry solute particle before condensation of water, and ρ_s is the density of the solute. The mass of water in the droplet is given by $m_w = \bar{v}_w \rho_w$, where $\bar{v}_w$ is the partial volume of water in the solution. In fact, since we are considering dilute solutions, we will approximate $\bar{v}_w \approx v = \frac{4}{3}\pi r^3$, where r is the droplet radius. Thus we may rewrite

$$\frac{e'}{e_s(\infty)} \approx 1 - \frac{\nu M_w a^3 \rho_s}{M_s r^3 \rho_w} = 1 - B \frac{a^3}{r^3}, \quad (1.15)$$

where

$$B = \frac{\nu M_w \rho_s}{M_s \rho_w} \quad (1.16)$$

is termed the Köhler hygroscopicity coefficient.

Köhler curves

For a droplet of solution the curvature and solute effects are combined such that

$$\frac{e_s(r)}{e_s(\infty)} = \left(1 - B \frac{a^3}{r^3}\right) \exp\left(\frac{A}{r}\right). \quad (1.17)$$

Expanding the exponential as a power series and defining supersaturation as $S = s - 1$, where $s = e_s(r)/e_s(\infty)$ is the saturation ratio at equilibrium, gives a simplified version of the Köhler equation (Rogers and Yau, 1989; Pruppacher and Klett, 1997; Seinfeld and Pandis, 1998):

$$S = \frac{e_s(r)}{e_s(\infty)} - 1 \approx \frac{A}{r} - B \frac{a^3}{r^3}. \quad (1.18)$$

Each individual Köhler curve is specific to the amount and composition of solute in the growing droplet (i.e. the dry radius, a) and composition of the initial aerosol particle. In Fig. 1.3, supersaturation over the droplet is plotted as a function of the increasing wet radius of the aqueous droplet, r , formed as the dry aerosol particle dissolves in the accumulating water.

At small radii the solute term dominates. As the droplet grows, the solution becomes more dilute and the Kelvin term dominates, which gives rise to the distinctive shape of the Köhler curve shown in Fig. 1.3(a) and also illustrated in Fig. 1.4. Differentiating supersaturation with respect to radius provides the location of the maximum of this curve at a critical supersaturation of

$$S_c = \frac{2}{\sqrt{B}} \left(\frac{A}{3a}\right)^{3/2}, \quad (1.19)$$

and critical radius of

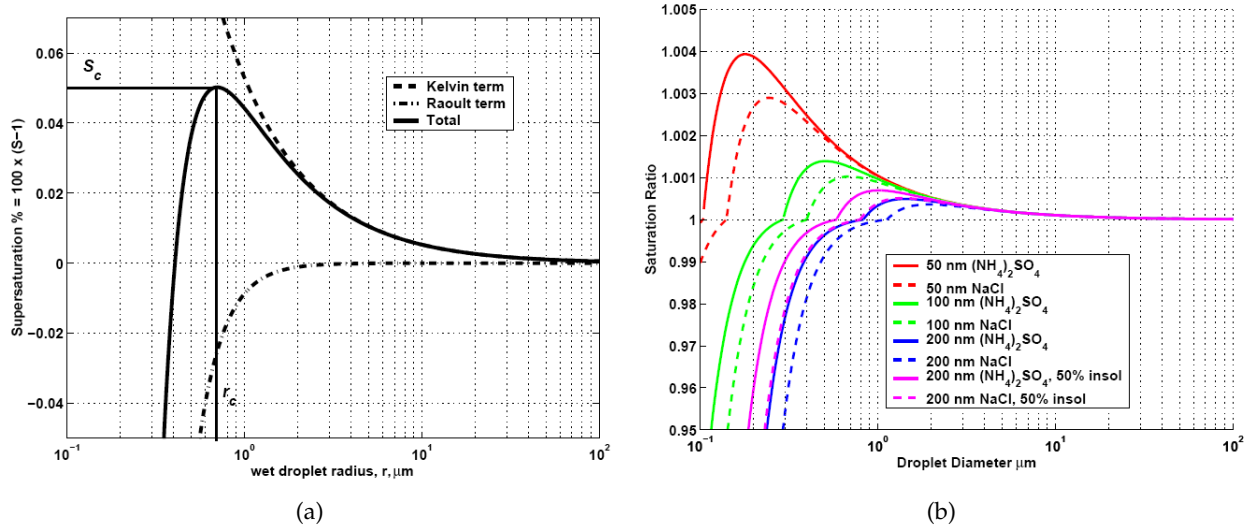


Figure 1.3: Competition between the curvature (Kelvin) and solute (Raoult) terms is shown in (a). The location of S_c and r_c are shown at the maximum of the curve. In (b) the dependence of activation on dry size is highlighted. Köhler (activation) curves are shown for a range of dry diameter of salt particles $(\text{NH}_4)_2\text{SO}_4$ with solid line, NaCl with dashed line). Red, green and blue lines indicate different dry diameters. Magenta lines are for a 200 nm particle containing 50% by mass insoluble core. Both from McFiggans et al. (2006).

$$r_c = \sqrt{\frac{3Ba^3}{A}}. \quad (1.20)$$

Beneath these critical values of supersaturation and radius, a droplet will be in stable equilibrium with the surroundings. If the relative humidity increases slightly, the droplet will grow slightly along the curve. Should the relative humidity decrease, some of the water in the droplet will evaporate until the vapour pressure over the droplet surface is in equilibrium with the ambient vapour pressure. If the relative humidity is increased to the critical supersaturation, the droplet will grow to the critical radius. Up to this critical point, an increase in droplet radius is dependent on an increase in ambient relative humidity. However, the significance of this critical point is that, beyond it, the Kelvin term dominates to such an extent that the saturated vapour pressure over the droplet decreases as the radius increases. Since the saturated vapour pressure over the droplet is less than the ambient vapour pressure, water will continue to condense onto the droplet and hence the droplet will continue to grow in a runaway process, given a sufficient source of water vapour (Köhler, 1936; Pruppacher and Klett, 1997; Seinfeld and Pandis, 1998; Wallace and Hobbs, 2006). These properties of the Köhler curve are illustrated in Fig. 1.4.

1.3.4 Aerosols as CCN

Both physical and chemical properties of an aerosol particle affect its ability to act as a cloud condensation nucleus (CCN), the most important being its size and solubility (as just discussed). Further chemical effects can also modify the propensity of an aerosol—particularly an organic aerosol—to act as a CCN. Ambient soluble gases, especially HNO_3 , NH_3 , and HCl , can dissolve in nascent droplets, increasing their soluble mass fraction, and

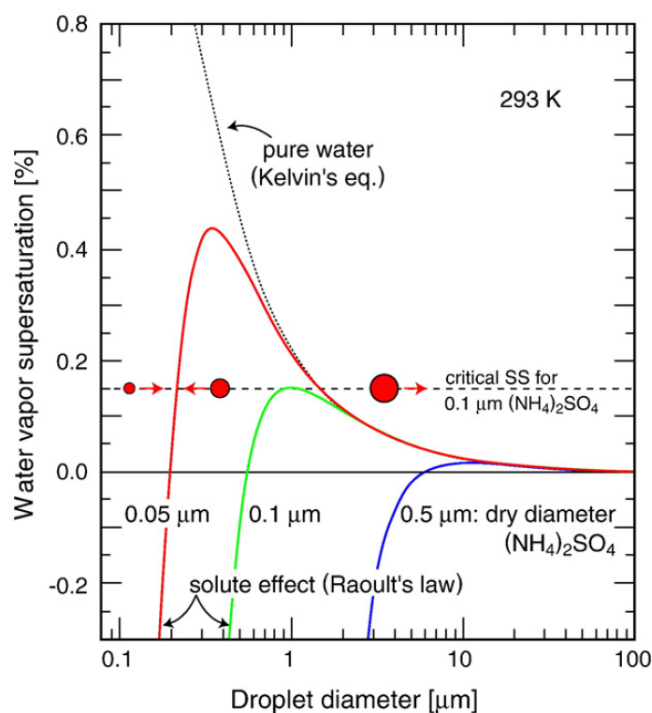


Figure 1.4: Köhler curves at 293 K for pure water (dotted line) and a range of dry diameter (denoted by colour) of $(\text{NH}_4)_2\text{SO}_4$ particles (solid lines). Dashed line indicates an ambient water vapour supersaturation, $S = 0.15\%$ which exceeds the critical supersaturation for $(\text{NH}_4)_2\text{SO}_4$ particles with dry diameter $\geq 0.1 \mu\text{m}$, so these aerosols will activate and grow into cloud droplets. Smaller particles remain as unactivated haze particles. Indicated drops (corresponding, for example, to a dry diameter of $0.05 \mu\text{m}$) show how these haze particles return to their equilibrium curve through condensation or evaporation. From Andreae and Rosenfeld (2008) after Seinfeld and Pandis (1998).

hence the rate of droplet growth (Kulmala et al., 1993; Hegg, 2000; Nenes et al., 2002). The presence of slightly soluble substances, especially organic compounds, can also increase the soluble mass fraction of the aerosol as relative humidity reaches 100% (Hartz et al., 2006). In the right conditions, even insoluble but non-hydrophobic aerosols can act as CCN due to the increase in droplet radius and dominance of the Kelvin term over the solute term at larger radius (Abbatt et al., 2005). Organic substances can reduce the surface tension in a droplet, aiding droplet growth (Shulman et al., 1996). However, organic constituents can also lead to the formation of a hydrophobic surface film, repelling water vapour and inhibiting activation or further growth (Kanakidou et al., 2005). A review of the nature and sources of cloud-active aerosols is given by Andreae and Rosenfeld (2008).

Observations and modelling studies indicate that after some time in the atmosphere, most aerosols are internally-mixed conglomerate particles, although the composition is not necessarily uniform across particular size classes (McFiggans et al., 2006). Currently however, in terms of modelling, variation in composition across a mode is difficult to represent. Instead, most models either assume a sum of uniform internally-mixed modes, or use a sectional representation of the size distribution, where the composition can be defined separately for each section.

1.3.5 Empirical relationships

The first attempts to represent and quantify aerosol–cloud interactions in GCMs relied on empirical relationships between cloud droplet number concentration (CDNC) and total aerosol mass (Boucher and Lohmann, 1995) or total aerosol number (Gultepe and Isaac, 1996; Jones et al., 1994). However, there is a large amount of uncertainty inherent in both stages of this method: uncertainty in relating aerosol and CCN number concentration to the aerosol mass concentration and uncertainty in relating CDNC concentration to CCN concentration (due to the complexities of aerosol activation as presented here). The complexity of aerosol–cloud interactions is such that a simple correlation based on a single aerosol property is bound to be an over-simplification. This is emphasised by a comparison of four different empirical relationships, including the two described, by Kiehl et al. (2000), yielding a very broad estimated range of the globally averaged radiative forcing due to the indirect aerosol effects from -0.40 W m^{-2} to -1.78 W m^{-2} .

1.4 Aerosol activation

A droplet that has grown to such a size that it exceeds its critical radius is said to be activated. Particles that can activate at a given supersaturation are defined as cloud condensation nuclei (CCN) for this supersaturation. Since activation is dependent upon the S_c of each particle being equalled or exceeded by the ambient supersaturation, the number of particles from a given aerosol population that can act as CCN is a function of the supersaturation.

Of course, such a droplet can only continue to grow beyond activation while there is still water vapour available to condense onto it. In the atmosphere there will usually be many particles competing for the available water vapour, and this provides a natural limit to the droplets' growth. The maximum supersaturation in a rising parcel of air occurs when the water vapour availability due to the parcel cooling as it rises balances the depletion rate of water vapour by those droplets that have already activated (Pruppacher and Klett, 1997). This relationship plays a fundamental role in modelling droplet formation. However, the kinetic limits imposed by the diffusion rate of water vapour also affect the rate at which water vapour may condense onto a population of droplets, which complicates matters beyond simply estimating the maximum supersaturation achieved in a cloud. These kinetic limitations are addressed in Sec. 1.4.3 and returned to subsequently in Chapter 2.

1.4.1 Converting an aerosol size distribution to a CCN spectrum

There are three distinct stages to modelling cloud droplet formation on aerosol in known atmospheric conditions. First, the aerosol distribution itself must be well-defined, in terms of its chemical composition and size distribution. Second, Köhler theory is required to re-express that size distribution in terms of the critical supersaturation needed to activate each of the aerosols in the distribution. This results in an expression for the number of droplets that could form as a function of supersaturation, known as the CCN spectrum. Third, the maximum supersaturation achieved in a parcel of air must be calculated. Disregarding any kinetic limitations for now, the CCN spectrum can then be integrated beneath $S_{\max}$ to calculate the total number of droplets that will form.

As described in Sec. 1.1.2, aerosol size distributions are commonly approximated as a power law, a sectional representation, or as multiple log-normal modes. HadGEM-UKCA has a multi-modal aerosol scheme. The aerosol activation parameterisation therefore requires the geometric mean radius and geometric standard deviation of each mode,

the chemical composition of the uniform internal mixture of each mode, and the number concentration of particles in each mode. Knowledge of the chemical composition of particles in each mode must include as a minimum the soluble mass fraction, degree of ionisation of soluble components, and the molecular mass of both soluble and insoluble components, but can also include information relating to additional chemical effects such as those described in Sec. 1.3.4.

Determining the maximum supersaturation in a rising parcel is essential for determining the number of aerosols that activate, since it follows from Köhler theory that the maximum supersaturation determines the radius of the smallest aerosol that activates, $a_{\min}$ (in a single mode of uniform composition). Evaluating Eq. (1.19) at $S_{\max}$ and rearranging gives,

$$a_{\min} = \frac{A}{3 \left(\frac{\sqrt{B}}{2} S_{\max} \right)^{2/3}}. \quad (1.21)$$

The number of activated aerosols in a mode can then be calculated by integrating the aerosol size distribution above this radius.

Alternatively, Von der Emde and Wacker (1993) showed that Köhler theory can be used to transform the log-normal number–size distribution, $dN/d \ln a$, to a number–supersaturation distribution and hence express the CCN spectrum in terms of log-normal parameters i.e.

$$\frac{dN}{d \ln S_c} = \frac{dN}{d \ln a} \frac{d \ln a}{d S_c}. \quad (1.22)$$

Differentiating Eq. (1.19) from Köhler theory gives

$$\frac{d \ln a}{d \ln S_c} = -\frac{2}{3}. \quad (1.23)$$

By rewriting Eq. (1.19) in terms of the critical supersaturation $S_{m,i}$ of a particle with the radius $a_{m,i}$ (the geometric mean radius of each aerosol mode, i) and combining this with Eq. (1.21), it follows that the size of the smallest aerosol that can activate can be expressed as

$$a_{\min} = a_{m,i} \left(\frac{S_{m,i}}{S_{\max}} \right)^{\frac{2}{3}}. \quad (1.24)$$

Substituting $dN/d \ln a$ from Eq. (1.5) for a multi-modal lognormal distribution, Eq. (1.23) and Eq. (1.24) into Eq. (1.22) allows the CCN spectrum to be written as

$$\frac{dN}{d \ln S_c} = - \sum_{i=1}^I \frac{2N_i}{3\sqrt{2\pi} \ln \sigma_i} \exp \left\{ - \frac{\ln^2 \left[(S_{m,i}/S_c)^{2/3} \right]}{2 \ln^2 \sigma_i} \right\}, \quad (1.25)$$

where $S_{m,i}$ is the critical supersaturation of a particle with the geometric mean radius of each aerosol mode, i . Eq. (1.25) can be integrated directly from 0 to $S_{\max}$ to give the total number of activated aerosols, N_a . With multiple log-normal modes, Von der Emde and Wacker (1993) show that the integral simplifies to³

$$N_a = \frac{1}{2} \sum_{i=1}^I N_i [1 - \operatorname{erf}(z_i)], \quad (1.26)$$

³The error function is defined as $\operatorname{erf}(x) = \frac{2}{\sqrt{\pi}} \int_0^x e^{-t^2} dt$.

where

$$z_i \equiv \frac{2 \ln (S_{m,i} / S_{\max})}{3\sqrt{2} \ln \sigma_i} = \frac{\ln (a_{\min} / a_{m,i})}{\sqrt{2} \ln \sigma_i}. \quad (1.27)$$

1.4.2 Maximum supersaturation

As an air parcel rises adiabatically it expands and cools. As it cools, the equilibrium vapour pressure of water vapour decreases [the temperature dependence of vapour pressure is given by the Clausius–Clapeyron expression, Eq. (1.7)], and since the total water content of the parcel stays constant, the supersaturation increases [see Eq. (1.18)]. The faster the parcel rises, the faster it cools and the greater the change in relative humidity with time. In terms of aerosol activation this is good news: greater updraught velocity and hence higher supersaturation in the parcel means a greater tendency for water vapour to condense onto the CCN and form cloud droplets. However, once activated, water vapour will continue to condense on the cloud droplets as they continue to grow, decreasing the supersaturation. The dry aerosol particles must then compete with already-activated droplets for the available water vapour. The balance between these two effects on the rate of change of supersaturation with time can be expressed as

$$\frac{dS}{dt} = \alpha w - \gamma \frac{dW}{dt}, \quad (1.28)$$

where w is the vertical updraught velocity of the parcel, $\frac{dW}{dt}$ is the rate of condensation of liquid water onto droplets, and α and γ are size-invariant coefficients (Pruppacher and Klett, 1997; Seinfeld and Pandis, 1998):

$$\alpha = \frac{gM_w L}{c_p R T^2} - \frac{gM_a}{RT}, \quad \gamma = \frac{pM_a}{e_s(\infty)M_w} - \frac{M_w L^2}{c_p R T^2}. \quad (1.29)$$

The maximum supersaturation $S_{\max}$ can be found when $\frac{dS}{dt} = 0$. The water condensation rate during activation is given by

$$\frac{dW}{dt} = 4\pi\rho_w \int_0^S r^2 \frac{dr}{dt} n(S') dS', \quad (1.30)$$

where $n(S')dS'$ is the number concentration of particles that activate between supersaturation S' and $S' + dS'$ and ρ_w is water density. We then substitute in the growth rate of a droplet, $\frac{dr}{dt}$, where r is the wet radius of the droplet (not to be confused with the dry radius of the original aerosol particle, a) (Pruppacher and Klett, 1997),

$$\frac{dr}{dt} = \frac{G}{r} \left(S - \frac{A}{r} + \frac{Ba^3}{r^3} \right), \quad (1.31)$$

where the growth coefficient, G , is size-dependent due to gas kinetic effects and defined as

$$G = \frac{1}{\frac{\rho_w R T}{e_s(\infty) D_v M_w} - \frac{L \rho_w}{K_a T} \left(\frac{L M_w}{R T} - 1 \right)}, \quad (1.32)$$

where K_a is the thermal conductivity of air and D_v is the water vapour diffusivity. Integrating Eq. (1.31) from the time τ when the parcel supersaturation equals the supersaturation required to activate a particle with the count median radius, to the time when maximum supersaturation is reached $t_{\max}$, gives the radius of the droplet at the time of $S_{\max}$

$$r^2(t) = r^2(\tau) + 2G \int_{\tau}^{t_{\max}} S(t) dt. \quad (1.33)$$

Substituting into Eq. (1.30) gives the water condensation rate as

$$\frac{dW}{dt} = 4\pi\rho_w GS \int \left(r^2(\tau) + 2G \int_{\tau}^{t_{\max}} S(t) dt \right)^{1/2} n(S') dS'. \quad (1.34)$$

Substituting this back into Eq. (1.28) with $\frac{dS}{dt} = 0$ provides the equation necessary to find $S_{\max}$:

$$\frac{\alpha w}{4\pi\gamma\rho_w} - GS_{\max} \int_0^{S_{\max}} \left(r^2(\tau) + 2G \int_{\tau}^{t_{\max}} S(t) dt \right)^{1/2} n(S') dS' = 0. \quad (1.35)$$

Lamentably, an analytical solution for the integral in this equation is not possible. Different models vary in the way they attempt to solve the integral numerically, from simple empirical fits to elaborate population-splitting iterative procedures. Solving this integral is the crux of the parameterisation of cloud droplet formation.

1.4.3 Kinetic limitations on droplet formation

Under equilibrium Köhler theory, introduced in Sec. 1.3.3, solving Eq. (1.35) to find $S_{\max}$, and then integrating the CCN spectrum below this value is sufficient to calculate the number of activated aerosols. Equilibrium Köhler theory assumes that aerosols are in equilibrium with the ambient supersaturation, and hence, in the case of increasing supersaturation, the particle grows gradually along its Köhler curve, in equilibrium with the surroundings until its critical supersaturation and radius are simultaneously reached, at which point it instantaneously activates.

In reality, the dynamic behaviour of a whole population of aerosols interacting with the ambient supersaturation requires several additional mechanisms to be accounted for. These are known collectively as kinetic limitations on droplet formation, and three key mechanisms have been identified (Nenes et al., 2001). The first mechanism concerns the activation of large aerosols that are termed ‘inertially-limited’. This inertial kinetic limitation arises when the timescale within which a particle would have to grow in order to remain in equilibrium with the increasing supersaturation of a rising parcel is shorter than the time it would actually take for that particle to grow by condensation (Chuang et al., 1997b). E.g. a particle with a large dry radius and very low critical supersaturation, $S_c < S_{\max}$ nevertheless may not have time to grow to its critical radius before $S_{\max}$ is reached, and thus would not actually activate, because condensational growth onto such particles is subject to kinetic limitations. Phinney et al. (2003) showed that falsely assuming such particles do activate, and grow to r_c (and beyond), leads to an overestimation of the amount of liquid water that they take up, which furthermore leads to a reduction in the amount of water vapour available to condense onto other non-inertially-limited droplets that could in fact activate within the timescale available (typically, these other non-inertially-limited droplets are smaller and more numerous than the inertially-limited droplets), and hence leads to an overall underestimation in droplet number.

At the other extreme, Barahona et al. (2010) showed that although such particles do not strictly activate, failure to account for the contribution to the condensational sink term provided by the large surface area of such inertially-limited particles leads to an

overestimation of $S_{\max}$, and hence an overestimation of the number of droplets that form. (This is demonstrated in Sec. 2.5.)

As the supersaturation in a parcel increases toward $S_{\max}$, the last particles to activate are usually the smallest aerosols, with the highest S_c . However, once the maximum supersaturation is reached, it may quickly decrease again, before runaway growth can take hold for these recently-activated aerosols with $S_c \approx S_{\max}$. Once S decreases below the equilibrium saturation ratio for such an aerosol, it deactivates, and returns to being interstitial aerosol, so this is known as the deactivation mechanism (Nenes et al., 2001). Due to the short amount of time for which $S_{\max}$ is maintained, larger particles with S_c slightly less than $S_{\max}$ may not be exposed to $S > S_c$ for long enough for them to activate at all, and so they simply evaporate to form interstitial aerosol without ever activating. This is known as the evaporation mechanism. Both the deactivation and evaporation mechanisms have been shown to have a significant effect on droplet number in highly polluted conditions, and should be taken account of in aerosol activation parameterisations (Nenes et al., 2001). Barahona et al. (2010) also demonstrated the need to account for the depletion of water vapour by condensation onto inertially limited particles, when giant CCN are present.

With these circumstances in mind, the assumption that aerosols with $S_c < S_{\max}$ will activate into droplets is sufficient for all other atmospheric conditions (Ghan et al., 2011), and thus the core of the parameterisation of aerosol activation is still the determination of $S_{\max}$ from Eq. (1.35).

1.4.4 Numerical models

While an analytical solution to Eq. (1.35) is not possible, there are various numerical aerosol activation models available that can explicitly calculate the time-evolution of an aerosol distribution in terms of the growth of particles by condensation and coagulation (Jensen and Charlson, 1984; Flossman et al., 1985). Most numerical models do this within the framework of an adiabatically-rising air parcel, but more detailed atmospheric conditions can be included in some models, such as incorporating the effects of entrainment of dry air into the parcel (Flossman et al., 1985).

There are two reasons why such numerical models are not currently incorporated into GCMs. Firstly the coarse spatial grid of a GCM does not have the level of detail required by such models. Secondly, the computational expense of running a GCM at the time resolution required by such models is prohibitive. At least twenty time-steps are typically required between initial saturation and maximum supersaturation (Ghan et al., 1993). Run at this resolution, the computation required by such detailed calculations would dominate the rest of the physics in most models.

One option for incorporating highly detailed models into GCMs is to run the numerical parcel model offline for a variety of different conditions and use a look-up table during the course of the atmospheric simulation. Unsurprisingly, this is only feasible when the activation process is only controlled by a few parameters. If the aerosol size distribution or composition are allowed to vary within the GCM, the size of look-up table required becomes impractical (Ghan et al., 2011).

1.4.5 Brief comparison of parameterisations

In order to consolidate the complexities of the microphysical aerosol activation process with the relatively coarse temporal and spatial resolution of a GCM, the activation process

must be parameterised in terms of the factors that influence it most significantly. Activation is a sub-grid-scale process. This means it acts on a smaller spatial scale than that of a GCM (typically $200 \times 200 \text{ km}^2$ horizontally and with ~ 20 vertical levels in the troposphere) and the process occurs over a time scale far briefer than a typical time step in a GCM ($\sim 20 \text{ min}$). Parameterising such processes means to express variability on smaller scales in terms of large-scale variables which are resolved in a GCM (Lohmann et al., 2007). A brief overview of the major developments in the field of aerosol activation parameterisation is given below, followed by a more detailed intercomparison of the behaviour of two of these parameterisations in Chapter 2. More detailed summaries and intercomparisons of the aerosol activation parameterisations currently in use in GCMs, can be found in the recent review paper by Ghan et al. (2011).

The parameterisation of activation must account for the competition between aerosol particles and for the dependence of this competition on particle size and distribution, and on the supersaturation forcing rate (i.e. updraught velocity). This approach relies on the parameterisation of other relevant sub-grid-scale processes within the GCM, in particular updraught velocity. Some GCMs apply a Gaussian distribution (Chuang et al., 1997a; Ghan et al., 2001b) or use turbulent kinetic energy as a proxy for the sub-grid-scale variability of updraught velocity (e.g. Lohmann et al. 1999). This topic is explored in more detail in Chapter 4. The parameterisation of sub-grid-scale cloud dynamics must be improved in tandem with the parameterisation of aerosol activation for useful, reliable results to be obtained within a GCM.

The first attempts to parameterise the number of cloud droplets that could form in an air parcel were made in the middle of the last century. Both Squires and Twomey developed approximate expressions for the number of droplets which would form in a uniform, adiabatically rising air parcel by estimating S_{max} and integrating an empirically-derived CCN spectrum (Squires, 1958; Twomey, 1959; Squires and Twomey, 1960). Squires makes the assumption that all CCN that become cloud droplets start growing from zero radius as soon as the parcel supersaturation exceeds zero. Twomey waits until the critical supersaturation of a CCN has been exceeded before growth begins, but then assumes that once this supersaturation is reached, that the CCN would instantaneously grow from zero radius to its final radius. Both methods empirically relate the number of activated droplets to supersaturation with a power-law relationship, $N = cS^k$ (where c and k are empirically-derived constants), but not to any fundamental properties of the aerosols on which the droplets form. The number of droplets is not even bounded by the total aerosol number and systematic errors arise, particularly at high updraught velocities or low aerosol number. Incidentally, it has been shown that this form of the supersaturation spectrum is consistent with a power law aerosol size distribution (Jiusto and Lala, 1981; Herbert, 1986). However, as discussed in section Sec. 1.1.2, there are serious limitations to the utility of such an approximation.

In recent years, Twomey's approach has been extended beyond the power-law size distribution to a more general expression that limits the number of activated aerosols at high supersaturation (Cohard et al., 1998). Khvorostyanov and Curry (2006) demonstrated that this was in fact an approximation to a log-normal distribution, which was later extended to the multi-mode case by Shipway and Abel (2010).

Ghan et al. (1993) use a single log-normal aerosol size distribution to derive an activation parameterisation which is naturally bounded at high updraught velocity. Köhler theory is used to transform this number–size distribution to a number–supersaturation distribution and hence express the CCN spectrum in terms of log-normal parameters. In contrast to the approach of Squires and Twomey, Ghan et al. neglect droplet growth beyond the point

where the supersaturation exceeds the critical supersaturation of each CCN, i.e. beyond $S > S_c$. Instead, it is assumed that the radius of each particle at $S = S_{\max}$ is given by its own critical radius, r_c . Clearly this is quite a simplification, since Köhler theory dictates that until $S = S_{\max}$, particles with $S_c < S_{\max}$ will experience spontaneous growth beyond r_c once $S = S_c$.

Abdul-Razzak et al. (1998) combine the approaches of Twomey (1959) and Ghan et al. (1993) by using a log-normal aerosol size distribution and including droplet growth between the time of activation of each particle and the time when $S = S_{\max}$. As shown in Sec. 1.4.2, approximate solutions to the set of simultaneous equations governing the time evolution of supersaturation in a rising air parcel [Eq. (1.28), Eq. (1.30), and Eq. (1.31)], are found by making the assumption that beyond activation, the curvature and solute effects do not strongly influence the rate of condensation of water onto activated droplets in Eq. (1.31), which is approximated as $\frac{dr}{dt} = \frac{GS}{r}$. This assumption allows Eq. (1.35) to be derived, which (as previously stated) has no analytic solution. Abdul-Razzak et al. used numerical simulations with a cloud parcel model to establish that an approximate solution to Eq. (1.35) could be found in terms of two dimensionless parameters, and two functions of the width of the aerosol size distribution, determined from empirical fits to the results from parcel model simulations in a wide range of input conditions.

More specific details are given in Chapter 2, in which the theory of the single log-normal aerosol mode is outlined (Sec. 2.2). This parameterisation has also been extended for the multi-mode case (Abdul-Razzak and Ghan, 2000), which is the formulation that is tested in Sec. 2.4.1 (albeit only employing one mode, for consistency with what will later be applied in the GCM). The multi-mode parameterisation is also extendable to the more general sectional representation of the aerosol size distribution, using many narrow log-normal modes (Abdul-Razzak and Ghan, 2002). The parameterisation has also been further extended to include the effects of organic surfactants which can suppress surface tension at the point of activation and increase the fraction of aerosols activated (Abdul-Razzak and Ghan, 2004).

Although the Abdul-Razzak and Ghan parameterisation performs well in a wide variety of conditions, one of its weaknesses is that it does not treat any of the kinetic limitations discussed in Sec. 1.4.3. It is also tuned to a fixed value of the condensation coefficient, $\alpha_c = 1.0$, which restricts the value of G [defined in Eq. (1.32)], although adjustments can be made to allow the inclusion of gas kinetic effects to some extent (Ghan et al., 2011).

Building on the work of Abdul-Razzak and Ghan, in a quest for a more general parameterisation with less dependence on empirical information, Nenes and Seinfeld (2003) developed an aerosol activation parameterisation that addresses some of the limitations mentioned above. In particular, a sophisticated ‘population splitting’ iterative approach allows for the explicit consideration of kinetic effects. The reliance on empirical information and correlations is substantially reduced, and eliminated altogether unless kinetic limitations are dominant. The concept of population splitting is explored in Sec. 2.3.2.

The Fountoukis and Nenes (2005) parameterisation (hereafter FN) extends the work of Nenes and Seinfeld (2003) by allowing for multi-mode log-normal aerosol size distributions (since these are prevalent in GCMs). It also addresses the limitations of Abdul-Razzak and Ghan (2000) by explicitly accounting for gas kinetic effects on water vapour diffusivity, dependent on aerosol size and condensation coefficient, which may be used to treat the film-forming ability of organic compounds. Barahona and Nenes (2007) extended FN beyond the adiabatic parcel by replacing α in Eq. (1.28) with a modified value that incorporates the effects of the entrainment of dry air into the parcel.

A more detailed description of the Abdul-Razzak and Ghan (2000) and Fountoukis and

Nenes (2005) parameterisations is given in Chapter 2 as well as an intercomparison of the behaviour of both schemes in controlled conditions. This analysis helped inform the choice of parameterisation for use in the GCM for the work in the rest of the thesis. It also led to a further development of the FN model, relating to the treatment of inertially-limited droplets (Barahona et al., 2010). This is described in Sec. 2.5.

Abdul-Razzak and Ghan (2000) and Fountoukis and Nenes (2005) are not the only parameterisations in use in current GCMs, but they are the most widespread. The next most prominent parameterisation is that of Ming et al. (2006). Briefly, this uses a sectional aerosol distribution, and carries out the integration required in Eq. (1.35) numerically. This parameterisation was not considered further for use in HadGEM-UKCA, which uses a multi-modal aerosol size distribution. Several alternative parameterisations have also been devised (e.g. Hänel 1987; Khvorostyanov and Curry 2008; Kivekäs et al. 2008; Khvorostyanov and Curry 2009; Shipway and Abel 2010), a summary of which can be found in Ghan et al. (2011). However, these were not considered as candidates for use in HadGEM-UKCA, either because they have not been demonstrated to compare so well against observations and numerical simulations as the two schemes that were considered (Abdul-Razzak and Ghan 2000 and Fountoukis and Nenes 2005), or because they were published after the choice had been made and technical work on the implementation had already begun. These approaches are therefore not addressed further in this work.

1.5 Methodology for modelling indirect aerosol effects

Once the number of activated aerosols has been calculated from the aerosol parameterisation in a GCM, this number is used to determine the cloud droplet number concentration (CDNC) in each cloudy model grid-box. Several other physical processes contribute to the indirect aerosol effects, and different models employ a variety of methods for parameterising these processes and their effects. Nevertheless, essentially, in order to model the indirect aerosol effects, this CDNC must be used in conjunction with the cloud liquid water content, to calculate the cloud droplet effective radius. This is an average cloud droplet size, used to determine the cloud albedo, and hence couple the influence of aerosols on clouds to the radiation scheme. Secondly, the number of droplets and cloud liquid water content are used to determine the rate by which cloud water is converted to rainwater. A generic summary of each of these processes is given below. More details of the specific implementation within HadGEM-UKCA are given in Chapter 3.

1.5.1 Cloud droplets from activated aerosols

First, the manner in which cloud droplet number concentration is estimated from the number of activated aerosols predicted by the activation scheme is dependent on the configuration of the cloud scheme within the host GCM.

Prognostic treatment of droplet number

The more physical approach, first implemented by Ghan et al. (1997), is to express conservation of droplet number as a differential equation which takes the general form,

$$\frac{\partial \overline{N_d}}{\partial t} = \frac{\partial \overline{N_d}}{\partial t} \Big|_{\text{adv}} + \frac{\partial \overline{N_d}}{\partial t} \Big|_{\text{mix}} + \frac{\partial \overline{N_d}}{\partial t} \Big|_{\text{activation}} + \frac{\partial \overline{N_d}}{\partial t} \Big|_{\text{sinks}}, \quad (1.36)$$

where $\overline{N_d}$ represents the grid-box mean cloud droplet number concentration. The terms on the right-hand side represent the changes in cloud droplet number due to, respectively: (1) advection, (2) mixing [both turbulent and convective], (3) aerosol activation [this term is a function of $\overline{N_a}$, the grid-box mean number of activated aerosols], (4) droplet sink terms [encompassing droplet evaporation, collision/coalescence, collection by rain, ice and snow, and freezing of water droplets to form ice crystals].

This approach is only available for models with a two-moment, prognostic cloud scheme, in which both number and mass of cloud droplets are stored as dynamically-varying quantities.

Diagnostic treatment of droplet number

Not all models have the necessary infrastructure (i.e. a dynamically evolving aerosol model coupled to a two-moment cloud-scheme) for N_d to be a prognostic quantity, and some must simply assume local equilibrium between the number of aerosols activated and the number of droplets that exist at any one time step, such that

$$N_d = f(\overline{N_a}). \quad (1.37)$$

Most such models that do this rely on a variety of empirically-derived relationships based on field measurements to calculate N_d directly from either the number or mass of modelled aerosol with no explicit dependence on w in the case of Boucher and Lohmann (1995) (and hence also by Dufresne et al. (2005)) and Jones et al. (2001). However, Chuang et al. (1997a) also treat droplet number diagnostically but do so with an aerosol activation scheme coupled to a pdf-based representation of cloud updraught velocity.

A more detailed review of the differing formulations used by existing GCMs, both prognostic and diagnostic, is given in the appendix of Golaz et al. (2011).

1.5.2 Effective radius calculation

Once the cloud droplet number concentration, N_d , has been determined, it is used in conjunction with the cloud liquid water content, q_c [$\text{kg}(\text{water}) \text{kg}(\text{air})^{-1}$], calculated by the model's cloud scheme, to generate the volume mean radius, r_v . This is related to a cloud droplet effective radius r_e , by β , which approximates the shape and spread of a cloud droplet size distribution:

$$r_e = \beta r_v = \beta \left(\frac{3}{4\pi} \frac{\rho_{\text{air}} q_c}{\rho_w N_d} \right)^{1/3}. \quad (1.38)$$

The dependence of cloud optical properties, such as cloud optical depth, single scattering albedo and asymmetry factor, on r_e , and their impact on the model's radiation scheme, allows the quantification of the radiative effects of changes to N_d on the Earth's radiation balance. This allows the cloud albedo effect to be modelled.

1.5.3 Autoconversion of cloud water to rain water

In order to calculate further indirect aerosol effects, a GCM must allow the cloud liquid water to respond to changes in cloud droplet number through the influence of CDNC on the autoconversion of cloud droplets to rain, which in turn influences the cloud lifetime (Ghan et al., 2001b). (Depending on local meteorological conditions, this can also affect the cloud height and cloud cover (Ackerman et al., 2004).) Any attempt to quantify the

uncertainties in modelled aerosol indirect effects must therefore include a comparison of the effects of different autoconversion schemes, which have been assessed to be a major source of uncertainty (Ghan et al., 2001b; Penner et al., 2006).

Lohmann and Feichter (2005) note that once autoconversion rate depends on the size or number of cloud droplets, the cloud lifetime effect can no longer be calculated separately from the first indirect aerosol effect without changing the reference state, but estimates of the separate effects can be obtained by prescribing a constant CDNC (Lohmann et al., 2000). This approach is used to estimate the separate contributions from each of the aerosol effects to the total in Chapter 6. Another commonly used approach which delivers a “quasi-” or “relaxed” forcing (Rotstayn and Penner, 2001; Penner et al., 2003) is to find the difference in net radiation at the top of the atmosphere for a present day and pre-industrial simulation using a GCM in atmosphere-only mode with fixed sea surface temperatures and sea-ice extent (Lohmann and Feichter, 2005; Jones et al., 2007). This later became known as a Radiative Flux Perturbation, and this is the method used to quantify and compare the indirect aerosol effects in different configurations of HadGEM-UKCA in Chapter 6.

1.5.4 Sources of uncertainty in estimates of the cloud albedo effect

A variety of GCMs have previously been used to make estimates of the radiative forcing due to the indirect aerosol effects, and many of these were included in the broad range of estimates provided by the Fourth Assessment Report of the IPCC (Forster et al., 2007). However, as shown by the IPCC in Fig. 1.2, there is a large amount of uncertainty in these estimates, and attributing the sources of these uncertainties is an important step towards reducing them. In their analysis of uncertainties in the cloud albedo effect, Chen and Penner (2005) considered the sources of uncertainty associated with:

- aerosol and aerosol precursor emissions,
- aerosol mass concentrations,
- aerosol size distributions,
- cloud droplet parameterisation,
- representation of in-cloud updraught velocity,
- the relationship between cloud droplet effective radius and volume mean radius,
- liquid water content,
- cloud fraction,
- change of cloud radiative properties due to the presence of black carbon.

This study used 3-D meteorological fields and a radiative transfer model to examine the spatially-resolved uncertainty due to each of these parameters in estimates of the first aerosol indirect effect. This study allowed the uncertain factors influencing the first indirect effect to be considered separately in a fair comparison, independent of host GCM, although it is of course limited by the choice of radiative transfer model. In some cases different parameterisations are compared, and in others extremes of the range of values found in the literature were used. The greatest source of uncertainty was found to be the aerosol mass concentration, which can increase forcing by up to -0.8 W m^{-2} . The second largest source

of uncertainty was attributed to the cloud fraction and thirdly the treatment of the cloud droplet spectral dispersion, β in Eq. (1.38).

Since the publication of this work by Chen and Penner (2005), concerted efforts have been made to address these dominant sources of uncertainty. In terms of estimates of the indirect aerosol effects with the GCM used in this thesis, several of the remaining large uncertainties have also been tackled in HadGEM3. The new prognostic cloud, prognostic concentrate (PC2) cloud-scheme in HadGEM3 (Wilson et al., 2008) specifically models prognostic cloud fraction, and thus the uncertainty due to cloud fraction should be substantially reduced. Finally, the introduction of the dynamically-evolving two-moment modal aerosol scheme in UKCA, which has been well-evaluated against measurements should also substantially reduce uncertainties in aerosol mass concentrations (and other properties) (Mann et al., 2010).

In the work which follows in this thesis, the choice has therefore been made to focus on the contribution of the representation of in-cloud vertical velocity in large-scale (stratiform) cloud to the uncertainty of the indirect aerosol effects, since this is the next most significant factor controlling cloud droplet number concentration, and hence indirect aerosol effects, assuming the uncertainties on the aerosols and clouds just described have already been reduced as far as possible. Updraught velocity plays a major role as an input to the aerosol activation parameterisations described in this chapter, two of which will be explored further in the next chapter. The parameterisation of sub-grid-scale vertical velocity is then considered in Chapter 4 and evaluated against measurements in Chapter 5. Finally the radiative effects of the choice of parameterisation, and hence a constraint on the overall uncertainty in estimates of the indirect aerosol effects arising from updraught velocity is given in Chapter 6.

Parameterisation of aerosol activation

Two of the aerosol activation parameterisations mentioned in the introduction, Abdul-Razzak and Ghan (2000) and Fountoukis and Nenes (2005), are discussed in more detail in this chapter. In particular, Sec. 2.2 and Sec. 2.3 describe their differing methods for determining the maximum supersaturation that is achieved in an adiabatically rising parcel, as $S_{\max}$ is the essential parameter that must be determined to predict the number of aerosols that activate. The behaviour of both schemes is examined in a box-model configuration, which allows specific areas of parameter space to be compared in a controlled fashion.

2.1 Calculation of maximum supersaturation

Once the log-normal aerosol size distribution has been converted to a CCN spectrum, as described in Sec. 1.4.1, all that remains is to examine the balance between sources and sinks of water vapour to determine the maximum supersaturation that occurs. Unfortunately, as shown in Sec. 1.4.2, the combination of the full set of governing equations leads to the following integral [repeated here from Eq. (1.35) for reference], which must be solved either approximately or iteratively in order to find $S_{\max}$:

$$\frac{\alpha w}{4\pi\gamma\rho_w} - GS_{\max} \int_0^{S_{\max}} \left(r^2(\tau) + 2G \int_{\tau}^{t_{\max}} S(t) dt \right)^{1/2} n(S') dS' = 0. \quad (2.1)$$

Each of these approaches is described below. These descriptions outline the approach when a single log-normal aerosol mode is used, although both schemes are capable of handling chemically complex aerosol with arbitrary external mixtures of multiple internally-mixed aerosol modes. The extension of the Abdul-Razzak et al. (1998) parameterisation to the multi-mode case is outlined in Sec. 2.2.1.

2.2 Abdul-Razzak and Ghan parameterisation

Abdul-Razzak et al. (1998) (and Abdul-Razzak and Ghan (2000)) examine the integral in Eq. (2.1) in two limits, which will be explored below. The parameter S_m is defined as the supersaturation required to activate a particle with the dry geometric mean radius, a_m . The size distribution is formulated in terms of the ratio $\frac{S_m}{S_{\max}}$ where $S_{\max}$ is again the maximum supersaturation attained in the parcel, and hence a proxy for the radius of the smallest particle that activates, $a_{\min}$ by Eq. (1.19).

In the first limit, the integral is considered for values of S_m small compared to $S_{\max}$ (i.e. where the dry radius of the smallest particle that activates, $a_{\min}$ is far smaller than the geometric mean radius of the mode, a_m). In this case the growth term dominates the r^2 term in Eq. (1.35). Once the negligible r^2 term has been removed, the resulting integral is solved using Twomey's lower bound (Twomey, 1959) to evaluate the inner integral. Substituting into Eq. (1.28) with $\frac{dS}{dt} = 0$ at $S_{\max}$ yields an expression for the upper bound of $S_{\max}$,

$$\frac{S_m}{S_{\max}} = S_m \sqrt{J / (\eta + 3\zeta)}, \quad (2.2)$$

where η and ζ are the dimensionless terms

$$\eta = \frac{(\alpha w G)^{3/2}}{2\pi\rho_w\gamma N}, \quad \zeta = \frac{2A}{3} \left(\frac{\alpha w}{G}\right)^{1/2}; \quad (2.3)$$

and J is defined as

$$J \cong \operatorname{erfc}(u) - \frac{1}{2} \left(\frac{S_m}{S_{\max}}\right)^2 \exp\left(\frac{3 \ln \sigma}{\sqrt{2}}\right)^2 \operatorname{erfc}\left(u + \frac{3 \ln \sigma}{\sqrt{2}}\right), \quad (2.4)$$

where

$$u \equiv \frac{\ln(a_{\min}/a_m)}{\sqrt{2} \ln \sigma} = \frac{2 \ln(S_m/S_{\max})}{3\sqrt{2} \ln \sigma}, \quad (2.5)$$

since $a_{\min}/a_m = (S_m/S_{\max})^{2/3}$.

In the limit that S_m is comparable to $S_{\max}$ (and $a_m \sim a_{\min}$), activated aerosols do not grow much beyond the point of activation. This is the limit proposed by Ghan et al. (1993). The growth term in the integral in Eq. (1.35) is negligible (as in the second limit in Sec. 2.3.1). Integrating the CCN spectrum, Eq. (1.25), and substituting into Eq. (1.28) with $\frac{dS}{dt} = 0$ at $S_{\max}$ gives

$$\frac{S_m}{S_{\max}} = U \frac{\zeta}{\eta}, \quad (2.6)$$

where

$$U \equiv \exp\left(\frac{3 \ln \sigma}{\sqrt{2}}\right)^2 \operatorname{erfc}\left(u - \frac{3 \ln \sigma}{\sqrt{2}}\right). \quad (2.7)$$

Thus far, the problem has been re-formulated, but not solved; both J and U are dependent on $S_{\max}$ and so an exact analytic solution is still impossible. Abdul-Razzak and Ghan propose an approximate solution based on numerical results of the full set of governing equations. These show that the ratio $(S_m/S_{\max})^2$ is proportional to $(\zeta/\eta)^{3/2}$ at small values of S_m and $(S_m/(\eta + 3\zeta))^{3/4}$ at large values of S_m . Over the full range of S_m the ratio is approximated by

$$\left(\frac{S_m}{S_{\max}}\right)^2 = f(\ln \sigma) \left(\frac{\zeta}{\eta}\right)^{3/2} + g(\ln \sigma) \left(\frac{S_m}{\eta + 3\zeta}\right)^{3/4}, \quad (2.8)$$

where J and U are replaced by f and g , functions of the geometric standard deviation of the aerosol size distribution. Abdul-Razzak et al. (1998) use the following expressions for these functions, again based on numerical simulations (they report an error of less than 25% for a realistic range over all governing parameters):

$$f(\ln \sigma) = 1.5 \exp\left(2.25 (\ln \sigma)^2\right); \quad g(\ln \sigma) = 1 + 0.25 \ln \sigma \quad (2.9)$$

Hence $S_{\max}$ is estimated from Eq. (2.8). As shown in Sec. 1.4.1, the CCN spectrum, Eq. (1.25), is then integrated below this value to give the total number of activated aerosols, N_a .

2.2.1 Extension to multiple log-normal modes

Abdul-Razzak and Ghan (2000) extended this parameterisation to treat the case of an external mixture of multiple log-normal aerosol modes, themselves each composed of an arbitrary internal mixture of chemical components, with all particles across all modes in competition with one-another for water vapour.

For multiple internally-mixed modes, the mean hygroscopicity coefficient averaged over all components $j = 1, 2, 3 \dots J$ of each mode i is calculated following Hänel (1976) and Pruppacher and Klett (1997) as

$$\overline{B}_i = \frac{M_w \sum_{j=1}^J x_{ij} \nu_{ij} \phi_{ij} \epsilon_{ij} / M_{sij}}{\rho_w \sum_{j=1}^J x_{ij} / \rho_{sij}}, \quad (2.10)$$

where ϕ_{ij} , x_{ij} and ϵ_{ij} are the osmotic coefficient, mass mixing ratio and soluble mass fraction, respectively, of component j and mode i . [The other symbols represent the same physical properties as they do in Eq. (1.16).]

The critical supersaturation required to activate an aerosol with the geometric mean radius in mode i can then be calculated, following Eq. (1.19),

$$S_{m,i} = \frac{2}{\sqrt{\overline{B}_i}} \left(\frac{A}{3a_{m,i}} \right)^{3/2}. \quad (2.11)$$

The equation for the rate of water condensation onto aerosols in multiple modes is generalised from Eq. (1.30) to give

$$\frac{dW}{dt} = 4\pi\rho_w \sum_{i=1}^I \int_0^S r_i^2 \frac{dr_i}{dt} n_i(S') dS', \quad (2.12)$$

where r_i is the radius of a droplet forming on the aerosol in mode i . The other necessary equations from the single-mode case are generalised in a similar fashion, with N , the total number of aerosols, being replaced by N_i , the total number in each mode, and summations being made over all I modes, where appropriate. Full details are given by Abdul-Razzak and Ghan (2000), but essentially, following the same method described above to derive Eq. (2.8) for the single mode case, a similar expression for $S_{\max}$ can be derived for the multimode case:

$$S_{\max} = 1 / \left\{ \sum_{i=1}^I \frac{1}{S_{m,i}} \left[f_i(\ln \sigma_i) \left(\frac{\xi}{\eta_i} \right)^{\frac{3}{2}} + g_i(\ln \sigma_i) \left(\frac{S_{m,i}}{\eta_i + 3\xi} \right)^{\frac{3}{4}} \right] \right\}^{\frac{1}{2}} \quad (2.13)$$

where $\eta_i = \frac{(\alpha w G)^{3/2}}{2\pi\rho_w \gamma N_i}$ and the functions f and g differ slightly from the single mode version:

$$f_i(\ln \sigma_i) = 0.5 \exp \left(2.5 (\ln \sigma_i)^2 \right); \quad g_i(\ln \sigma_i) = 1 + 0.25 \ln \sigma_i. \quad (2.14)$$

These updated functions reflect a retuning of the parameterisation against numerical simulations with a baseline aerosol size distribution more appropriate for the accumulation

mode particles that largely determine the total number activated¹ (Abdul-Razzak and Ghan, 2000). As described above for the single-mode case, once $S_{\max}$ has been determined, the total number of activated aerosols, N_a , can be calculated as shown in Sec. 1.4.1, using Eq. (1.26),

$$N_a = \frac{1}{2} \sum_{i=1}^I N_i [1 - \operatorname{erf}(z_i)]$$

where

$$z_i \equiv \frac{2 \ln(S_{m,i}/S_{\max})}{3\sqrt{2} \ln \sigma_i},$$

and where Eq. (2.11) has been used to determine $S_{m,i}$ for each mode. The fraction of activation, $N_{\text{act}} = N_a / \sum_{i=1}^I N_i$ can then be calculated trivially.

2.3 Nenes *et al.* parameterisations

Nenes and Seinfeld (2003) introduced the concept of population splitting to allow for the explicit consideration of kinetic effects on aerosol activation. As a result, the reliance on empirical information and correlations is substantially reduced compared to the Abdul-Razzak and Ghan parameterisation, and eliminated altogether in a number of conditions. This method is outlined below in Sec. 2.3.2. Fountoukis and Nenes (2005) details the extension to the case of an external mixture of multiple internally mixed log-normal aerosol modes, (following a similar method to the one described above for the extension to the Abdul-Razzak and Ghan parameterisation) and this is the formulation that is tested in Sec. 2.5.

2.3.1 Asymptotic limits in the Nenes *et al.* models

Nenes and Seinfeld (2003) use a sophisticated bisection method to solve the integral in Eq. (1.34) by iteration. Following the notation in the paper (using diameter, D , instead of radius, r), the integral alone is

$$I(0, S_{\max}) = GS_{\max} \int_0^{S_{\max}} \left(D_p^2(\tau) + 2G \int_{\tau}^{t_{\max}} S(t) dt \right)^{1/2} n(S') dS', \quad (2.15)$$

where $D_p(\tau)$ is the diameter of a CCN when it is exposed to the critical supersaturation, S_c , needed for it to activate, and τ is the amount of time taken to reach that critical supersaturation. Nenes and Seinfeld consider the two asymptotic limits of $I(0, S_{\max})$ by comparing the two terms in the integral. In the first limit the CCN continue to grow significantly beyond the point where the ambient supersaturation equals their own critical supersaturation,

$$D_p^2(\tau) \ll 2G \int_{\tau}^{t_{\max}} S(t) dt. \quad (2.16)$$

That is, the size at which the CCN activates, $D_p(\tau)$, is much less than the size it will finally grow to by the time $S = S_{\max}$. In this case, Twomey's lower bound (Twomey, 1959) can be used to approximate the supersaturation integral, as described in Sec. 1.4.5:

¹Note that, in the (single-mode) tests that follow, the functions derived for the multi-mode case are used, since these would be used in a GCM with multiple aerosol modes.

$$\int_{\tau}^{t_{\max}} S(t) dt \approx \frac{1}{2\alpha w} (S_{\max} - S(\tau)^2), \quad (2.17)$$

where $S(\tau)$ is the parcel supersaturation at time τ . Substituting back gives the integral in the first limit

$$I_1(0, S_{\max}) = GS_{\max} \int_0^{S_{\max}} \left(\frac{1}{2\alpha w} (S_{\max} - S(\tau)^2) \right)^{1/2} n(S') dS'. \quad (2.18)$$

The second limit occurs when the CCN do not grow significantly beyond the point where the ambient supersaturation exceeds their own critical supersaturation. (This is the limit employed by Ghan *et al.* (1993), in contrast to the choices of Squires (1958); Squires and Twomey (1960) and Twomey (1959).) I.e.

$$D_p^2(\tau) \gg 2G \int_{\tau}^{t_{\max}} S(t) dt. \quad (2.19)$$

For the smallest CCN that activates, there is no time for growth beyond the critical size, $D_p(\tau)$ (or r_c). In this case the growth rate, $\frac{dr}{dt}$, is zero, so from Eq. (1.31),

$$S_c = \frac{A}{r_c} - B \left(\frac{a}{r_c} \right)^3. \quad (2.20)$$

Substituting for the expression for r_c , Eq. (1.20), yields²

$$r_c = \frac{2A}{3S_c}. \quad (2.21)$$

In the context of the Nenes *et al.* parameterisations, $D_p(\tau)$ is assumed to be equal to³ $D_c = 2A/3S_c$ when the parcel supersaturation, S , equals the CCN critical supersaturation, S_c . This is a reasonable assumption when $\frac{dr}{dt} \ll \frac{dS}{dt}$ (Chuang *et al.*, 1997a). It assumes that a CCN is in equilibrium with the varying parcel supersaturation, and hence it reaches the critical radius simultaneously with the supersaturation reaching the CCN's critical supersaturation.

Substituting the approximation of Eq. (2.19) into Eq. (2.15) yields

$$I_2(S_c, S_{\max}) = \frac{2}{3} AGS_{\max} \int_{S_c}^{S_{\max}} \frac{n(S')}{S_c} dS'. \quad (2.22)$$

2.3.2 Population Splitting

The two limits of the integral in Eq. (2.15) described above apply to CCN with contrasting attributes. The concept behind population splitting is that the aerosol distribution can be divided into two groups; CCN which are best described by the approximations made for Eq. (2.18) and CCN which are more appropriately described by the approximations underlying Eq. (2.22). In supersaturation space, these two groups are divided by the partition supersaturation, S_{part} . The integral, $I(0, S_{\max})$ in Eq. (2.15) is approximated by

$$I(0, S_{\max}) = I_1(0, S_{\text{part}}) + I_2(S_{\text{part}}, S_{\max}). \quad (2.23)$$

²Note that this is not an approximation, but an exact analytical expression (Ghan *et al.*, 1993).

³Nenes and Seinfeld (2003) define the Köhler A coefficient as twice the value defined in equation 1.12, which accounts for the factor of two between diameter and radius which appears to be missing here.

We assume there is only one S_{part} and that the upper bound of I_1 and the lower bound of I_2 coincide. Those CCN that would follow $I_1(0, S_{\text{part}})$ are those with either $D_p \ll D_c$ or $D_p \gg D_c$ and grow significantly beyond $S > S_c$. Those which do not grow significantly beyond $S = S_c$ are those with $D_p \approx D_c$ and follow $I_2(S_{\text{part}}, S_{\text{max}})$. The method for determining S_{part} is described in detail in Nenes and Seinfeld (2003) and a brief outline follows.

The first choice for S_{part} is the CCN population critical supersaturation, such that $D_p^2(\tau) = 2G \int_{\tau}^{t_{\text{max}}} S(t) dt$. From Eq. (2.17), and substituting $D_p(\tau) = 2A/3S_{\text{part}}$, this eventually generates the quartic equation

$$S_{\text{part}}^4 - S_{\text{max}}^2 S_{\text{part}}^2 + \frac{4A^2 \alpha w}{9G} = 0. \quad (2.24)$$

There are two real roots to this equation if the discriminant, $\Delta = S_{\text{max}}^4 - \frac{16A^2 \alpha V}{9G}$, is non-negative

$$\left(\frac{S_{\text{part},1}}{S_{\text{max}}} \right)^2 = \frac{1}{2} \left[1 - \left(1 - \frac{16A^2 \alpha w}{9S_{\text{max}}^4 G} \right)^{1/2} \right]; \quad \left(\frac{S_{\text{part},2}}{S_{\text{max}}} \right)^2 = \frac{1}{2} \left[1 + \left(1 - \frac{16A^2 \alpha w}{9S_{\text{max}}^4 G} \right)^{1/2} \right]. \quad (2.25)$$

These roots describe the behaviour of CCN that do not grow significantly beyond $S = S_c$, are those with $D_p \approx D_c$ and follow $I_2(S_{\text{part}}, S_{\text{max}})$. The second root is chosen in the model. When the discriminant is negative, Eq. (2.24) has no real roots. In this case S_{part} is calculated using a correlation derived from the regression of a large set of empirical S_{part} that reproduce the predictions of a detailed microphysics model:

$$\frac{S_{\text{part}}}{S_{\text{max}}} = \min \left(0.666 \times 10^7 A S_{\text{max}}^{-0.3824}, 1.0 \right). \quad (2.26)$$

Once S_{part} has been determined, Eqns. 2.18 and 2.22 are substituted into Eq. (2.23), which approximates Eq. (2.15) and replaces the integral in Eq. (1.34). This can be solved for S_{max} using an iterative bisection method (see algorithm in Fig. 2.1). The final form of the parameterisation is

$$\frac{\pi}{2} \frac{\gamma \rho_w G S_{\text{max}}}{\rho_{\text{air}} \alpha w} \left(\int_0^{S_{\text{part}}} D_p n^s(S') dS' + \int_{S_{\text{part}}}^{S_{\text{max}}} D_p n^s(S') dS' \right) - 1 = 0 \quad (2.27)$$

$$\frac{\pi}{2} \frac{\gamma \rho_w G S_{\text{max}}}{\rho_{\text{air}} \alpha w} [I_1(0, S_{\text{part}}) + I_2(S_{\text{part}}, S_{\text{max}})] - 1 = 0. \quad (2.28)$$

The discriminant, Δ , from Eq. (2.24) (which can be seen in the algorithm schematic in Fig. 2.1), expresses the extent of kinetic limitations throughout the droplet formation and depends on S_{max} , w and the timescale of water vapour diffusivity (embodied in G). $\Delta = 0$ marks the boundary between two growth regimes. For $\Delta > 0$, most CCN are free from kinetic limitations and S_{part} is determined without empirical information using the second root shown in Eq. (2.25). For $\Delta < 0$, kinetic limitations in droplet growth are strong throughout the entire population and S_{part} is calculated using the empirical relationship shown in Eq. (2.26), which is based on numerical simulations which were chosen so that $\Delta < 0$. This is the only part of the parameterisation which relies on empirical information, and it is required for approximately 20% of cases (Nenes and Seinfeld, 2003; Fountoukis and Nenes, 2005).

Once S_{max} has been determined, the number of activated aerosols can be calculated by integrating the CCN spectrum below this value in exactly the same way as described above for the Abdul-Razzak and Ghan parameterisation.

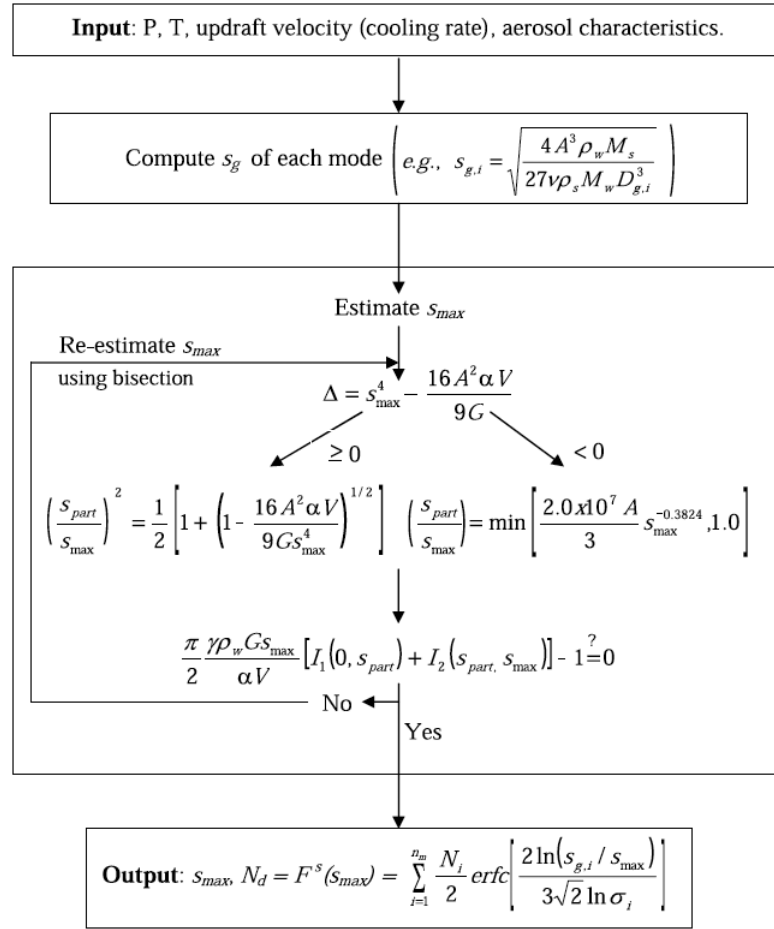


Figure 2.1: Schematic representation of the parameterisation algorithm for the log-normal formulation (FN). From Fountoukis and Nenes (2005).

2.4 Parameterisation intercomparison

In order to choose which parameterisation to implement in HadGEM-UKCA, it was decided to perform a study to explore and compare the behaviour of these two schemes in a zero-dimensional box-model setup (effectively at a single model grid-point). Both parameterisations are able to treat multi-modal aerosol distributions, but the tests which follow were performed with just a single aerosol mode, for clarity. In particular, this study focused on how the two models behave as a function of updraught velocity and particle size, although number concentration and soluble mass fraction were also investigated. Hereafter, the Abdul-Razzak and Ghan (2000) aerosol activation parameterisation is referred to as 'ARG', and the Fountoukis and Nenes (2005) parameterisation as 'FN'.

2.4.1 Abdul-Razzak and Ghan parameterisation

The ARG box model has been used to examine how the fraction of activated aerosol particles varies with updraught velocity, w and the count median diameter of a single log-normal

Aerosol number concentration (m^{-3})	230×10^6
Geometric standard deviation (dimensionless)	1.47
Density of soluble fraction (kg m^{-3})	1760.0
Density of insoluble fraction (kg m^{-3})	2100.0
Soluble mass fraction (dimensionless)	0.33
Molar mass of soluble fraction (kg mol^{-1})	1.32×10^{-1}
van 't Hoff factor for soluble fraction (ions molec $^{-1}$)	3.0

Table 2.1: Input parameters defining North Atlantic marine aerosol test case, provided by Nenes.

aerosol mode⁴, d_m (dry diameter). Updraught velocity was varied over the range 0.001 to 10 m s^{-1} and d_m was varied from 0.005 to $10 \mu\text{m}$ at a temperature $T = 298 \text{ K}$ and pressure $p = 900 \text{ mbar}$. Aerosol composition and size distribution follow the marine aerosol type described in Table 2.1. Since both parameters extend over a range of orders of magnitude, incremental steps are spaced logarithmically to avoid over-sampling high values. The model was run using combinations of 100 values of d_m and 100 values of w .

Fig. 2.2 shows results from these runs of the model at three important stages. As described in Sec. 2.2, the ARG parameterisation is used to determine the maximum supersaturation achieved at a grid-point, $S_{\max}$. This is shown in Fig. 2.2 (a) as a function of both w and d_m . As expected, the contours show that $S_{\max}$ depends strongly on both parameters. In the adiabatic approximation, as w increases, so does $-dT/dt$ and hence the supersaturation increases. With increasing particle size, the Kelvin effect ensures that saturated vapour pressure decreases over the increasingly-flatter surface of larger particles with a larger radius of curvature. Hence condensation is more favourable, resulting in a decrease in $S_{\max}$, as can be seen in the diagram.

Köhler theory relates $S_{\max}$ to the critical size of the smallest particle that can activate, $d_{\min}$, since it follows from Eq. (1.19) that $S_{\max} \propto d_{\min}^{-3/2}$. Fig. 2.2 (b), is simply a direct translation of (a) in which colour and black contour lines denote $d_{\min}$ (rather than $S_{\max}$) as a function of w and d_m .

Finally, the aerosol size distribution is integrated above $d_{\min}$ to obtain the total number of activated aerosols, N_a . This is plotted in Fig. 2.2 (c) as a fraction of the total number of aerosols in the mode, known as the fraction of activation, N_{act} . (Note that when count median diameter, $d_m = d_{\min}$, 50% of particles are activated. This is clearly shown in (b) at intersections of black contours of $d_{\min}$ with white dashed vertical lines of constant d_m and the white solid line of 50% activation. This gives some confirmation that the integration step of the algorithm is reliable.)

Up to a dry diameter of $\sim 0.07 \mu\text{m}$, N_{act} increases with both increasing updraught velocity and increasing count median diameter of the aerosol mode. At very low updraught velocities, for very small particles, there is no activation. As the size distribution is shifted along to higher values of d_m and the updraught velocity is increased, the fraction of activated particles increases. Beyond $\sim 0.2 \mu\text{m}$, the aerosols are of such a size that increasing the median value of the distribution no longer has a noticeable effect on the fraction of activation, and the updraught velocity is the limiting factor on maximum supersaturation,

⁴N.B. For a log-normal distribution, the median and the geometric mean are coincident. The phrase ‘count’ (or ‘number’) median diameter is used to distinguish it from the ‘mass’ median diameter. Diameter is frequently used instead of radius in aerosol science, although radius has been used as much as possible up to here for clarity and familiarity in equations.

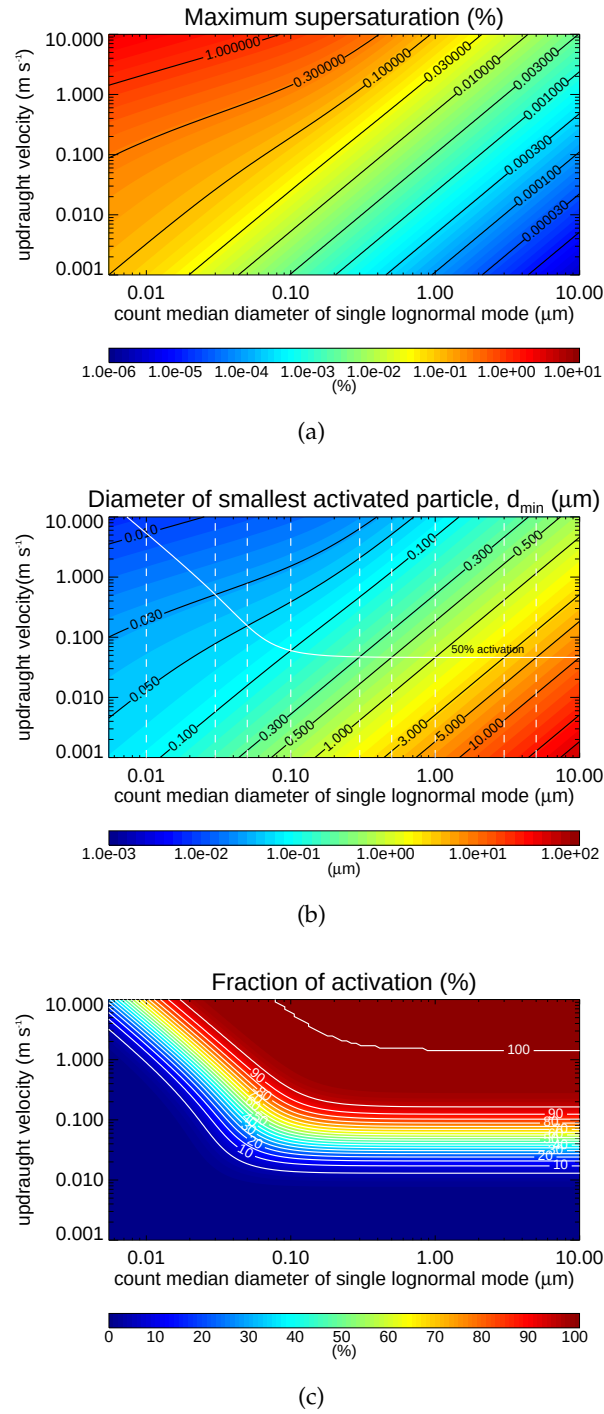


Figure 2.2: Results from ARG box model, run over a range of w and d_m for the marine aerosol test case (detailed in Table 2.1). In (a), colour shows $S_{\max}$ (%). In (b), colour and black contour lines show $d_{\min}$ (μm). White dashed lines show that the white solid line of $N_{\text{act}} = 50\%$ occurs when $d_{\min} = d_m$, as expected. In (c), colour shows fraction of activation, N_{act} .

and hence N_{act} .

2.5 Fountoukis and Nenes parameterisation development

The same tests were performed with the Fountoukis and Nenes (2005) parameterisation, in a similar box-model configuration, under the same conditions as described for ARG, again examining how N_{act} varies with dry particle modal count median diameter and updraught velocity. However, the FN scheme was found to behave unexpectedly in regions of simultaneous low updraught velocity and large count median diameter, as shown compared to ARG in Fig. 2.3 (a–d). In the FN parameterisation, the fraction of activation of aerosols under these conditions is 100%, compared to less than 5% in the ARG model. In addition, there is a discontinuity in the first derivative of the contours of constant fraction of activation (and hence a similar discontinuity in the first derivative in values of fraction of activation in slices of constant w or d_m) for the FN parameterisation.

These box model studies paved the way for collaborative development between the author, and Nenes and Barahona, and led to the eventual publication of an updated version of the FN parameterisation, FN2 (Barahona et al. 2010). Initially, the author raised concerns over the results shown in Fig. 2.3 (a–d). These two-dimensional contour plots make clear the problem of excess activation for large count median particle diameter. This results from an overestimation of S_{max} at large particle size and low updraught velocity, and substantiated concerns about the treatment of inertially-limited particles in the original FN parameterisation. Barahona and Nenes then proposed a potential solution that provided the framework for the final formulation. However, this preliminary version involved an unnecessary stage which actually circumvented population splitting altogether over the entire parameter space. This was demonstrated theoretically and confirmed through further box-model simulations by the author. Several discrepancies between the mathematical formulation in the draft paper and the code were also identified. The unnecessary stage was removed by Barahona *et al.*, who then performed further detailed comparisons between this updated version of the parameterisation and a numerical parcel model (shown later in Fig. 2.4). These comparisons show considerable improvements compared to the original FN scheme, and resulted in the final formulation, published in Barahona, West, Stier, Romakkaniemi, Kokkola, and Nenes (2010).

In this paper, we propose that the unexpected overestimation of N_{act} , for large d_m at low w [shown in Fig. 2.3 (d) compared with (b)], is due to a failure to account for the contribution to the condensational sink provided by inertially-limited droplets. Here, ‘inertially-limited’ refers to aerosols with a large dry diameter and hence very low critical supersaturation, which nevertheless cannot grow to their critical diameter in the timescale typically required to reach S_{max} and hence cannot be considered strictly ‘activated’. However, the liquid water content and surface area of such droplets may be comparable to that of strictly activated particles, and so neglecting to account for this surface area leads to an underestimation in the condensational sink term, in the equation for the rate of change of supersaturation with time [i.e. the second term in Eq. (1.28): $\frac{dS}{dt} = \alpha w - \gamma \frac{dW}{dt}$], which is fundamental to the determination of S_{max} .

In the updated scheme, dW/dt is modified to account for the effects of these large CCN, and is re-written as the sum of two terms: the first, the standard rate of condensation onto strictly activated particles, computed using the standard FN parameterisation, dW_{ps}/dt , and the second, the rate of condensation onto the inertially-limited particles, dW_{ie}/dt . I.e. when both activated and inertially-limited CCN contribute to the condensation sink, then

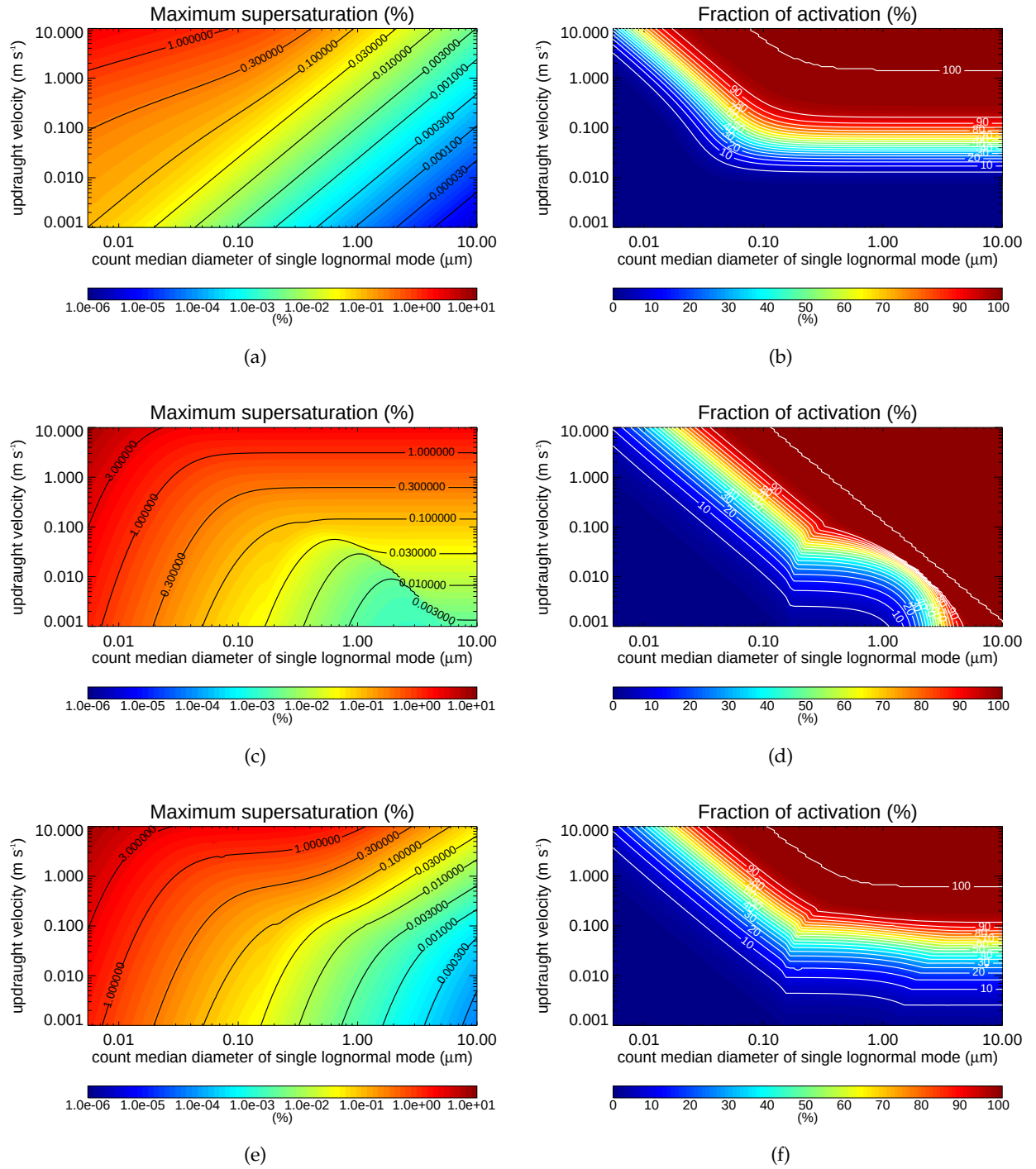


Figure 2.3: Results of box-model studies of three different parameterisations of aerosol activation. In LH column, colour contours show maximum supersaturation, $S_{\max}$ as a percentage, and in the RH column, fraction of activation, N_{act} , calculated for 100 values over each of the given ranges of updraught velocity and count median diameter for the marine aerosol test case (Table 2.1). (a) and (b) show the ARG parameterisation, (c) and (d) show the FN parameterisation and (e) and (f) show the updated FN2 model, which accounts for inertial limitations (Barahona et al. 2010).

dW/dt at the point of $S_{\max}$ is the sum of these two terms:

$$\frac{dW}{dt} = \frac{dW_{ie}}{dt} + \frac{dW_{ps}}{dt}. \quad (2.29)$$

Due to the large initial size of inertially-limited CCN, they do not substantially increase in wet diameter between cloud base and the level of $S_{\max}$. The condensational sink can be calculated by first considering the amount of liquid water at cloud base, in equilibrium with these inertially-limited aerosols:

$$W_{ie} = \frac{\pi \rho_w}{6 \rho_{air}} \left[\int_{D_p^{\min}}^{\infty} D_p^3 n(\ln D_p) d \ln D_p \right], \quad (2.30)$$

where $n(D_p)$ is the droplet size distribution, and D_p is their equilibrium size at saturation (when $S = 0$). The lower limit on the integral, $D_p^{\min}$, is the equilibrium diameter at saturation of the smallest particle that could activate (i.e. $D_p^{\min}$ is the size at $S = 0$ of a particle with $S_c = S_{\max}$). This lower limit can be related to $S_{\max}$ via Köhler theory as $D_p^{\min} = \frac{2}{3\sqrt{3}} \frac{A}{S_{\max}}$.

Assuming the growth rate of these droplets is given by $dD_p/dt = GS/D_p$ (Nenes and Seinfeld 2003), where G is the growth coefficient defined in Eq. (1.32), but that the diameter D_p does not in fact change between cloud base and $S_{\max}$, differentiating Eq. (2.30) with respect to time gives

$$\frac{dW_{ie}}{dt} = \frac{\pi \rho_w}{2 \rho_{air}} GS_{\max} \left[\int_{D_p^{\min}}^{\infty} D_p n(\ln D_p) d \ln D_p \right]. \quad (2.31)$$

This can be re-written as⁵

$$\frac{dW_{ie}}{dt} = \frac{\pi \rho_w}{2 \rho_{air}} GS_{\max} N_a \bar{D}_p, \quad (2.32)$$

in terms of $\bar{D}_p$, the mean droplet diameter (assuming an entirely inertially-limited size distribution), defined as

$$\bar{D}_p = \frac{1}{N_d} \int_{D_p^{\min}}^{\infty} D_p n(\ln D_p) d \ln D_p, \quad (2.33)$$

where N_d is the total droplet number concentration [i.e. $N_d = \int_{D_p^{\min}}^{\infty} n(D_p) dD_p$] and $n(\ln D_p) = dN_d/d \ln D_p$.

When both activated and inertially-limited CCN contribute to condensation, then Eq. (2.32) can be substituted back in to dW/dt [in Eq. (2.29)] and therefore Eq. (2.28), the final equation in the original FN algorithm shown in Fig. 2.1, is replaced by

$$\frac{\pi \gamma \rho_w GS_{\max}}{2 \rho_{air} \alpha w} \{ I_1(0, S_{part}) + I_2(S_{part}, S_{\max}) + N_a \bar{D}_p \} - 1 = 0. \quad (2.34)$$

This is solved iteratively to find $S_{\max}$, and hence the number of activated aerosols.

The behaviour of this updated parameterisation, FN2, was evaluated by Barahona *et al.*, who performed simulations with a numerical cloud parcel model (Nenes *et al.*, 2001)

⁵The full derivation of this parameterisation can be found in Barahona *et al.* (2010)

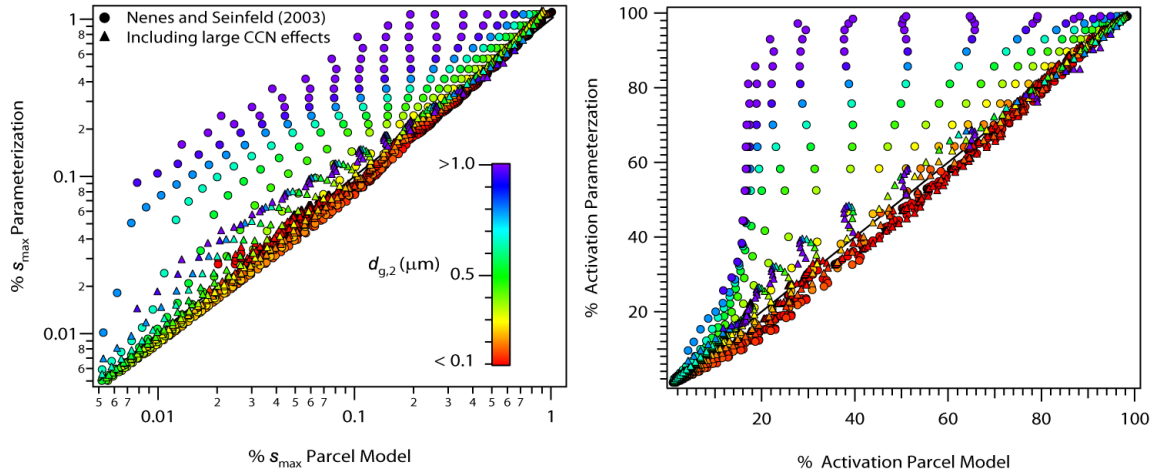


Figure 2.4: Maximum supersaturation (left) and aerosol activation fraction (right) for a bimodal aerosol distribution. Results are from application of the Nenes and Seinfeld (2003) parameterization neglecting (circles) and considering (triangles) the effect of kinetic limitations on large CCN. Conditions considered are $N_{\text{conc},1} = 2000 \text{ cm}^{-3}$, $N_{\text{conc},2} = 400 \text{ cm}^{-3}$, $\sigma_{g,1} = \sigma_{g,2} = 1.59$, $d_{m,1} = 0.08 \mu\text{m}$, $T = 290 \text{ K}$, $p = 100 \text{ kPa}$ and $\alpha_c = 0.06$. Colour coding varies with the count median diameter of the second mode. Symbols within the same colour vary with updraught velocity. Reproduced from Fig. 1 of Barahona et al. (2010).

over a wide range of updraught velocities and aerosol size distribution characteristics. The predicted S_{max} and N_{act} were compared with those from both the original and updated parameterisations.

In these simulations, the aerosol is assumed to be pure ammonium sulphate made up of two lognormal modes, with $N_{\text{conc},1} = 2000 \text{ cm}^{-3}$, and $N_{\text{conc},2} = 400 \text{ cm}^{-3}$ respectively. The geometric dispersion of both modes is set to $\sigma_{g,1} = \sigma_{g,2} = 1.59$ and $d_{m,1} = 0.08 \mu\text{m}$. (This aerosol was chosen to be representative of atmospheric conditions, described by Seinfeld and Pandis (1998)). Updraught velocity was varied over the range 0.01 to 10 m s^{-1} and the size of the second mode varied from $d_{m,2} = 0.005 \mu\text{m}$ to $5 \mu\text{m}$. In Fig. 2.4, colour indicates the size of the second mode of aerosols $d_{m,2}$. Results from the original parameterisation, FN, (without the correction due to inertial limits), compared to the parcel model, are shown as circles in Fig. 2.4. It is clear that this parameterisation significantly overestimates S_{max} and N_{act} compared to the parcel model, particularly for $d_{m,2} > 0.5 \mu\text{m}$ (green and blue circles). Triangular symbols show the results from the updated parameterisation, FN2. The introduction of the correction due to inertially limited particles substantially improves the agreement between the parameterisation and the parcel model, in the conditions tested, as can be seen by comparing the locations of the triangles to the circles in Fig. 2.4.

The results of running this updated version of the parameterisation, FN2, in a box model, (under exactly the same configuration as for the original model) are shown in Fig. 2.3 (e) and (f). The proposed modification does successfully eliminate the overestimation of supersaturation caused by the underestimation in surface area from inertially-limited particles. However, the abrupt (and unphysical) discontinuities in the first derivative of contour lines of N_{act} and S_{max} in the region around $0.1 \leq d_m \leq 0.4 \mu\text{m}$ still persist. Further box-model simulations indicate that this ‘kink’ occurs where the expression used to derive S_{part} changes, apparently due to the influence of kinetic limitations on droplet activation (Ghan et al. 2011). To a lesser extent there is also a second ‘kink’ in the $2 \leq d_m \leq 4 \mu\text{m}$

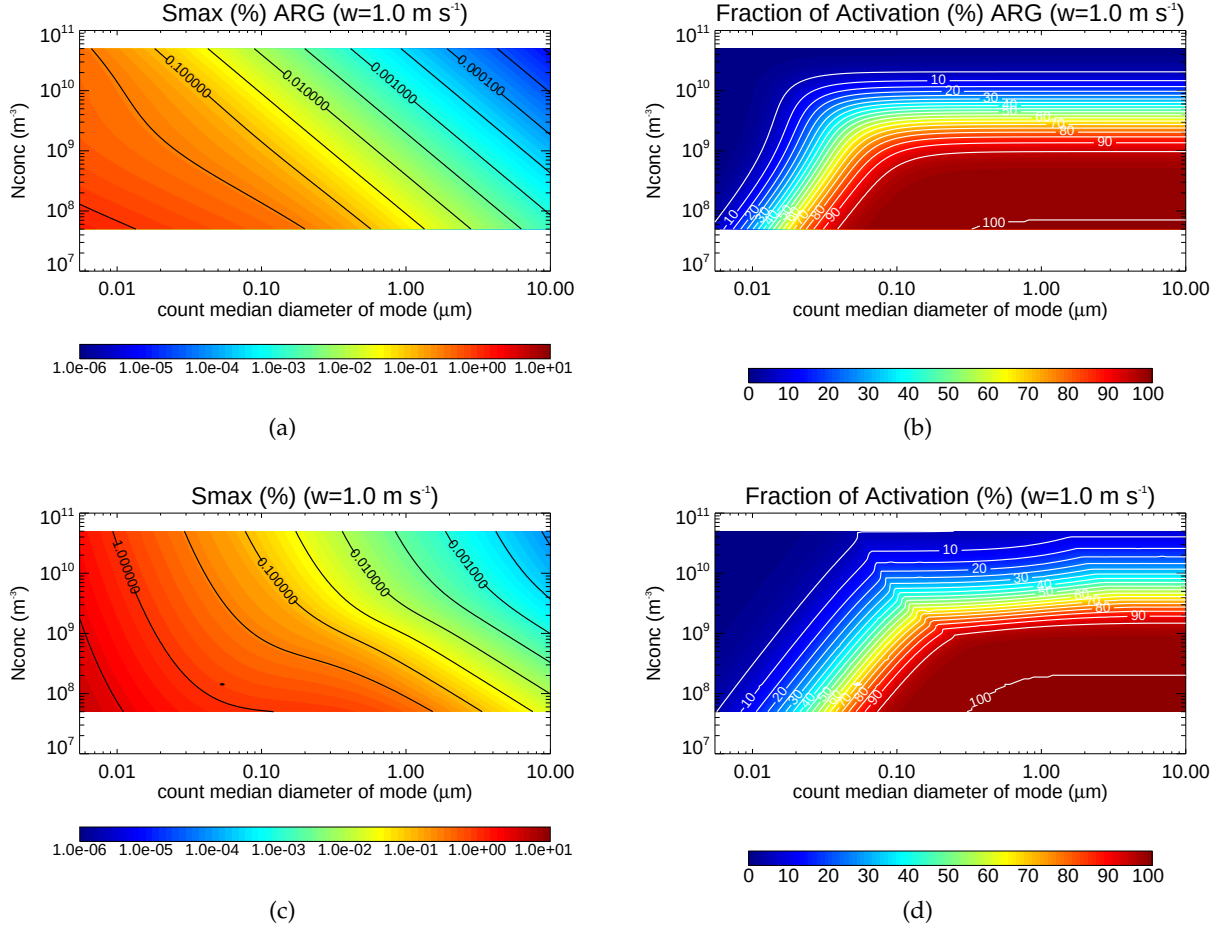


Figure 2.5: Colour denotes S_{max} in figures (a,c) and N_{act} , in figures (b,d) calculated over a range of aerosol number concentration and d_m for the marine aerosol test case (detailed in Table 2.1), with a fixed updraught velocity of 1 m s^{-1} . (a) and (b) show ARG parameterisation while (c) and (d) show FN2 parameterisation.

range. This is due to the transition to the regime where inertially-limited particles begin to dominate. Additionally, Fig. 2.5 demonstrates that both of these kinks persist in another area of parameter space, where N_{act} and S_{max} are plotted as a function of the initial number concentration of aerosols, N_{conc} and particle size, d_m . (In these simulations, updraught velocity was fixed at $w = 1 \text{ m s}^{-1}$.) Future work on the Nenes family of parameterisations will be devoted to smoothing out these transitions, to minimise the potential impact of such anomalous step changes in regions of parameter space which may have relevance to GCM studies.

2.6 Choice of parameterisation for HadGEM-UKCA

The timescale of the work in this thesis was such that it was necessary to begin the technical work of implementing an aerosol activation parameterisation into HadGEM-UKCA (the GCM) before the issues regarding the treatment of inertially limited particles in the Nenes model had been fully resolved. The contrast between the results of the initial

box-model studies [i.e. Fig. 2.3 (b) and (d)] therefore left little choice but to implement the Abdul-Razzak and Ghan scheme. Weaknesses of this scheme have been highlighted in particular by Phinney et al. (2003), who found that its lack of ability to account for inertial limitations means that (in contrast to the FN scheme) it tends to overestimate the amount of water taken up by inertially limited droplets, and hence underestimate the total number of droplets forming. They found this to be particularly true for aerosol concentrations $N_{\text{conc}} > 500 \text{ cm}^{-3}$.

In a recent comprehensive intercomparison between the four parameterisations most commonly used in current GCMs (Abdul-Razzak and Ghan (2000); Barahona et al. (2010); Ming et al. (2006); Shipway and Abel (2010)), Ghan et al. (2011) found that the more complex parameterisations (such as FN2) perform well under the widest range of conditions, but that all four parameterisations perform well under the most common atmospheric conditions, and predicted cloud droplet number concentrations to within 30% of those found with a detailed numerical model. Consistent differences were found between all four schemes, and these were largest for the Ming et al. (2006) scheme. Since the ARG scheme will be used in GCM studies culminating in radiative forcing calculations it is worth bearing in mind the strengths and weaknesses of the scheme itself when compared to a numerical cloud parcel model. The findings of Ghan et al. with regards to ARG are therefore summarised here.

Compared to a numerical parcel model, in a single mode case, ARG is found to underestimate S_{max} for updraughts stronger than 1 m s^{-1} . When the updraught is fixed at 0.5 m s^{-1} , ARG consistently underestimates S_{max} by about 10% over a wide range of aerosol number concentration, and predicts S_{max} remarkably well with varying aerosol size and width of size distribution and composition (tested by varying the hygroscopicity parameter), demonstrating the utility of empirical fits in some circumstances.

Intercomparisons with the parcel model were also carried out with several tri-modal aerosol test cases of different aerosol types (from Whitby 1978) over a range of updraught velocity from $w = 0.1$ to 10 m s^{-1} . The weaknesses of the ARG scheme become more apparent when it is required to deal with these more complex cases. For the marine aerosol case, it is found to underestimate the maximum supersaturation by 40% for all w , leading to an underestimation of activation in the nucleation mode in strong updraughts and underestimates in the accumulation mode in weak and moderate updraughts. For clean continental aerosol, these biases are weaker. For background aerosol conditions, ARG has been shown to underestimate S_{max} and N_{act} by about 30% across the range of updraughts. Finally, in the urban aerosol case, ARG was also found to underestimate S_{max} across the range of updraughts compared to the parcel model, although it performs slightly better than FN2 for updraughts stronger than 1 m s^{-1} . This underestimation of S_{max} leads to an underestimation compared to the numerical simulation of N_{act} in the accumulation mode for strong updraughts and, in the coarse mode for weak updraughts.

An underestimation of S_{max} leads to an underestimation of the number of aerosols that activate, and hence an underestimation in N_d . To speculate on how this might affect estimates of the indirect effect it is necessary to consider how N_d could change in simulations with both pre-industrial and present-day aerosol distributions. Consider a region where the number of CCN is markedly higher in the PI than PD cases. Imagine that the underestimation of S_{max} leads to the same relative underestimation in N_d for both the PI and PD cases. Then the decrease in absolute N_d due to underestimating S_{max} in the PD case would be greater than the decrease in N_d in the PI case, simply because the CCN concentrations are so much higher in the PD scenario. This would lead to a smaller contrast in N_d between the PI and PD cases, a smaller change in cloud albedo and hence an underestimation of the indirect effect. I.e. if the underestimation in S_{max} leads to a

reduction of N_d by some fraction in both PI than PD cases then ΔN_d between the two runs, and hence the indirect aerosol effects, will also be underestimated.

There are some situations where the ARG parameterisation performs better than FN2, usually because it has been tuned to the numerical simulations, although in most cases the FN2 model performs better because it is based on more robust physics. In particular ARG predicts lower $S_{\max}$ and N_{act} than FN2 in low aerosol concentrations, and more activation than FN2 in high aerosol concentrations, such that it is more sensitive to perturbations to aerosol number concentration, most likely because of its inability to deal with the kinetic limitations discussed in Sec. 1.4.3.

Ghan et al. (2011) also performed an intercomparison of the behaviour of both FN2 and ARG in GCM simulations with a GCM (CAM5). They find that the difference between present-day and pre-industrial shortwave cloud forcing is -1.60 W m^{-2} with FN2 and -1.76 W m^{-2} with ARG, so the ARG scheme may lead to higher estimates of the indirect aerosol effects, which should be borne in mind in later chapters.

2.7 Summary and conclusions

In this chapter, the different parameterisations of $S_{\max}$ used by the ARG and FN schemes have been compared in controlled box-model simulations and the response of each model to variations in updraught velocity, number and size of aerosols was investigated. As expected, both models showed that $S_{\max}$ increases as a function of updraught velocity, and decreases as a function of aerosol size and number concentration, although the relationships are non-linear. The fraction of activation behaves in a similar way, although obviously it is naturally bounded never to exceed unity in each mode. However, the FN model was found to behave unexpectedly in regions of low updraught velocity and large aerosol size. This was found to be due to an underestimation of the water-vapour depletion by inertially-limited aerosols, leading to an overestimation of $S_{\max}$ and hence N_{act} in this region. Collaborative work between the author, Barahona and Nenes led to the development of an updated version of the FN parameterisation that incorporates the effect of large inertially-limited particles. Accounting for these effects leads to substantial improvements in the agreement between the updated parameterisation, FN2 and a numerical cloud parcel model, when compared to the original FN scheme. This work was published as Barahona, West, Stier, Romakkaniemi, Kokkola, and Nenes (2010).

The timescale for the development of the updated FN2 scheme was such that implementation of the ARG scheme into HadGEM-UKCA had already begun by the time that the developments were finished and evaluated against the parcel model. At the time of implementation the ARG appeared to be a more reliable choice of parameterisation, but intercomparisons by Ghan et al. (2011) have shown that FN2 performs consistently well across a wide range of multi-dimensional parameter-space. The physical basis of FN2 is robust, and the concept of population splitting allows it to explicitly model the different kinetic limitations that affect aerosol across the size ranges. Nevertheless, improvements are still needed to smooth the transitions between regimes which lead to unphysical discontinuities in the first derivative of $S_{\max}$ and hence N_{act} , which occur in climatically-relevant regions of parameter space, and thus may have implications in GCM studies.

UKCA-Activate: incorporating microphysical aerosol–cloud interactions into HadGEM-UKCA

This chapter describes the implementation of the Abdul-Razzak and Ghan aerosol activation scheme within the Hadley Centre Global Environment Model, HadGEM3 (Hewitt et al., 2011). This model has been coupled to a new framework for modelling atmospheric chemistry and aerosols, known as UKCA (United Kingdom Chemistry and Aerosols community model), which has been recently developed in a collaborative effort between the UK Met Office and the Universities of Leeds, Cambridge and Oxford. The chapter gives a brief overview of HadGEM and UKCA before focusing on the implementation of the aerosol activation scheme and its interactions with other aspects of HadGEM. The ability of UKCA to produce suitable cloud condensation nuclei is evaluated by comparing modelled cloud condensation nuclei (CCN) number concentrations with measurements from 55 separate observational studies. Finally, a qualitative evaluation of several modelled cloud and aerosol properties against data from Earth-observing satellites is presented.

3.1 Introduction to HadGEM-UKCA

The UKCA model is coupled to HadGEM3, the latest incarnation of the Met Office Unified Model (UM). Full details of the HadGEM3 family of models are given by Hewitt et al. (2011). As a climate modelling system, HadGEM3 contains major developments from HadGEM1 and HadGEM2, most importantly the coupling of new ocean and sea-ice components to the UM atmosphere. This forms the physical basis for the Earth System model HadGEM3-ES. However, in this thesis, all integrations of the model involve the atmosphere-only configuration, HadGEM3-A, in which sea-surface temperatures and sea-ice extent are prescribed as seasonally varying fields, so no further details are given of the ocean and sea-ice developments.

HadGEM3-A is available with a variety of resolutions, and here the horizontal latitude-longitude grid used is a staggered Arakawa C-grid (Arakawa and Lamb, 1977) with a resolution of N96 (1.25° latitude $\times$ 1.875° longitude). A staggered Charney–Phillips grid is used in the vertical with 38 levels extending up to 39 km. The dynamical timestep corresponding to this resolution is 30 minutes.

Coupled within the climate model, UKCA uses components of HadGEM3 for the large-scale advection, convective transport, and boundary layer mixing of its chemical tracers. The large-scale transport is based on the new dynamical core implemented in HadGEM by Davies et al. (2005). Advection is semi-Lagrangian with conservative and monotone treatment of tracers. Convective transport is treated according to the mass-flux scheme of Gregory and Rowntree (1990) and is applicable to moist convection of all types (shallow, deep, and mid-level) in addition to dry convection. For boundary layer mixing, UKCA uses a new boundary layer turbulent mixing scheme (Lock et al., 2000) which includes a representation of non-local mixing in unstable layers and an explicit entrainment parameterisation.

To emphasise that the configuration of HadGEM3 used in this thesis is HadGEM3-A coupled to the UKCA aerosol and chemistry schemes, the model is hereafter referred to as HadGEM-UKCA.

3.1.1 Summary of chemistry component of UKCA

The UKCA chemistry scheme is available in either troposphere-only, stratosphere-only or combined (troposphere plus stratosphere) configurations. For all the model simulations in this thesis, UKCA is used with the standard tropospheric chemistry scheme, coupled to the aerosol model GLOMAP-mode (see below). The scheme simulates the O_x , HO_x and NO_x chemical cycles and the oxidation of CO, ethane and propane with eight emitted species (NO_x , CH_4 , CO, HCHO, C_2H_6 , C_3H_8 , CH_3COCH_3 and CH_3CHO) tracked by 27 prognostic tracers, 102 gas-phase reactions, 27 photolysis reactions and interactive wet and dry deposition schemes (Abraham et al., 2012).

Sulphur chemistry is coupled to the GLOMAP-mode aerosol scheme (described below) by extending the standard tropospheric scheme to treat the degradation of the aerosol pre-cursor species: sulphur dioxide (SO_2), dimethylsulphide (DMS), and monoterpene. In addition, two tracers are used to represent species required for the processes of aerosol nucleation and condensation within GLOMAP-mode: sulphuric acid (H_2SO_4) produced from the oxidation of SO_2 with the hydroxyl radical (OH), and a secondary organic species representing the condensable species from monoterpene oxidation.

A scheme for the oxidation of SO_2 within clouds by dissolved hydrogen peroxide (H_2O_2) and ozone (O_3) is also provided to give sulphate production rates which are used for in-cloud aerosol growth rates in GLOMAP-mode.

3.1.2 Overview of aerosol component of UKCA: GLOMAP-mode

Aerosol modelling in UKCA is accomplished with a new version of the Global Model of Aerosol Processes (GLOMAP), itself an extension of the TOMCAT global 3-D offline chemical transport model (Chipperfield, 2006). GLOMAP-mode is a two-moment pseudo-modal scheme which carries both aerosol number concentration and component mass as prognostic tracers. This allows processes such as condensation of gases on to aerosol particles to be represented by increasing the mass in a particular size class (mode), while conserving particle number.

The aerosol dynamics framework follows that of the M7 model (Vignati et al., 2004). Details of the aerosol modes and the permitted component species within each mode are given in Table 3.1. Mineral dust was not available as a component of GLOMAP-mode within UKCA at the time that the simulations used in this thesis were performed. Instead, dust is treated by the existing HadGEM 6-bin dust scheme (Woodward, 2001). (This aspect

Mode	Soluble?	Diameter range (μm)	Composition	σ_g
Nucleation	Yes	$\bar{D} < 0.01$	SO ₄ , OC	1.59
Aitken	Yes	$0.01 < \bar{D} < 0.1$	SO ₄ , BC, OC	1.59
Accumulation	Yes	$0.1 < \bar{D} < 1$	SO ₄ , BC, OC, SS, [DU]	1.59
Coarse	Yes	$\bar{D} > 1$	SO ₄ , BC, OC, SS, [DU]	2.0
Aitken	No	$0.01 < \bar{D} < 0.1$	BC, OC	1.59
Accumulation	No	$0.1 < \bar{D} < 1$	[DU]	1.59
Coarse	No	$\bar{D} > 1$	[DU]	2.0

Table 3.1: Configuration of the GLOMAP-mode aerosol scheme, with component species: sulphate (SO₄), sea salt (SS), black carbon (BC), organic carbon (OC), and dust (DU). Note that dust is not currently available for use in UKCA. σ_g is the geometric standard deviation of each log-normal aerosol mode.

of UKCA is still under development, as is the inclusion of ammonium nitrate as an aerosol component.) The M7 framework is referred to as pseudo-modal because the process rates for each mode are based on a particle size given by a single diameter, rather than by integrating over the full size range for each mode. This simplification is applied in several modal aerosol schemes [e.g. Wilson et al. 2001; Vignati et al. 2004; Stier et al. 2005].

Within GLOMAP-mode, aerosol processes are represented following the approach of the original sectional aerosol model, GLOMAP-bin (Spracklen, 2005; Spracklen et al., 2008). This includes nucleation of sulphuric acid, condensation, coagulation and cloud processing. Direct size-resolved emissions of sulphate, black carbon, organic carbon and sea salt particles are included, and secondary aerosol production from sulphur and terpene oxidation (prescribed at a fixed rate) is taken into account. Hygroscopic growth of all modes and ageing of insoluble modes by condensation or coagulation with soluble components are included. Finally, aerosol removal by both dry and wet deposition (sedimentation and scavenging) are also included. Full details of these processes are comprehensively described by Mann et al. (2010).

3.1.3 Coupling of aerosols and chemistry to the HadGEM3 radiation scheme

Aerosols and chemistry are both coupled directly to the HadGEM3 radiation scheme. Radiatively active trace gases simulated by the chemistry scheme can affect radiative fluxes at every timestep. The direct interaction of aerosols with radiation, by scattering and absorption of radiative fluxes, is controlled by the RADAER scheme. Full details are given by Bellouin (2010) but, essentially, aerosol optical properties are derived from some of the dynamically-varying aerosol properties simulated by GLOMAP-mode, such as the aerosol mass, number concentration, mean radius, and the amount of aerosol water in the soluble modes. The refractive index of each mode varies with the changing internal composition of the mode, and must also be calculated interactively. The aerosol optical properties (i.e. the specific scattering and absorption coefficients, and the asymmetry parameter, which describes the angular dependence of scattering) can then be calculated online at each radiation timestep, and the interactions of aerosol with both long- and short-wave radiation can be modelled. Aerosol optical depth¹ diagnostics at several wavelengths are also generated by RADAER.

¹Aerosol optical depth is the total extinction at a given wavelength due to aerosol in an atmospheric column.

3.2 Implementation of aerosol activation parameterisation: UKCA-Activate

Of course, aerosols can also interact indirectly with radiation through their effects on clouds. As described in Chapter 1, one of the primary aims of the work in this thesis is to implement an aerosol activation parameterisation into HadGEM-UKCA, to allow this process to be modelled. Aerosol activation is critically dependent on the number, size and composition of aerosols, as well as the local supersaturation of water vapour. The UKCA activation scheme, referred to as UKCA-Activate, has been designed to explicitly represent these factors by coupling the dynamically-evolving two-moment-modal aerosol scheme GLOMAP-mode to a Köhler theory based aerosol activation parameterisation, Abdul-Razzak and Ghan (2000).

The following section details how the Abdul-Razzak and Ghan aerosol activation scheme (described and tested in the previous chapter) has been implemented within HadGEM-UKCA, such that aerosol simulated by UKCA can be used to diagnose cloud droplet number concentration, which is then used in the radiation code and large scale precipitation scheme to simulate the indirect aerosol effects.

In this work we only consider the effect of aerosols on clouds, but not vice versa (other than a simplistic representation of scavenging), due to limitations in the current configurations of both UKCA and the HadGEM3 cloud scheme, PC2. In the PC2 scheme (Wilson et al., 2008), cloud fraction and cloud water content are both prognostic variables. However, PC2 is a single-moment cloud scheme, which means that while mass of cloud water is tracked, the number concentration of cloud droplets is not a prognostic variable, and is not stored from one timestep to the next. At every timestep, the cloud droplet number concentration is simply diagnosed afresh, directly from the number of aerosol particles available in each grid-box at that time, with no memory of how many cloud droplets there were at the previous timestep, or how many aerosols were previously activated.

This restricts the lifetime of cloud droplets to the timestep of the model, in this case 30 minutes. As it happens, the lifetime of cloud droplets is of this order, which is one reason why diagnostic droplet number is a reasonable approximation in terms of cloud properties.

However, the implications of diagnostic cloud droplet number for aerosol are that UKCA effectively ignores most cloud processing of aerosol. In the absence of a two-moment cloud scheme, it is impossible to track the number of aerosol particles that are contained within cloud droplets (known as in-cloud aerosol), although the mass fraction of in-cloud aerosol is estimated by UKCA. Secondly, any interactions between cloud droplets which could affect the number of aerosol particles (e.g. by coalescence or subsequent evaporation of droplets) are overlooked. The mass fraction of in-cloud aerosol can be estimated from the cloud fraction and liquid water content, which is used to estimate the in-cloud oxidation of SO_2 and subsequent increases in aerosol mass, but it would be preferable to treat the number of in-cloud, interstitial, and out-of-cloud aerosols as three separately evolving populations, since they are subject to different microphysical processes.

The act of diagnosing N_d purely from the number of aerosols that activate is structurally different to a prognostic scheme for cloud droplet number. A prognostic scheme carries forward pre-existing droplets, and considers a number of other sources and sinks of N_d (such as via aerosol activation, advection, mixing, evaporation, collision/coalescence, sedimentation and freezing). A diagnostic scheme does not bring forward N_d , but attempts to encapsulate the range of source and sink processes by (re)activating aerosols afresh at each timestep (i.e. droplets which would have been carried forward in a prognostic scheme are reactivated at each timestep). Whether this results in a systematic bias in estimates of

N_d , or a local one in different regions of parameter space, has been investigated by Ghan et al. (1997), amongst others, and a similar investigation will be performed with HadGEM3 with the forthcoming introduction of a prognostic cloud droplet number scheme.

Fortunately, future developments of HadGEM are set to include new two-moment cloud microphysics, with a prognostic equation for cloud droplet number concentration. UKCA should take advantage of this by incorporating explicit treatment of in-cloud aerosol to allow more realistic cloud–aerosol interactions to be modelled.

3.2.1 Inputs to UKCA-Activate

UKCA-Activate uses the Abdul-Razzak and Ghan (2000) aerosol activation scheme, which predicts the number of activated aerosols, N_a , as a function of aerosol properties (size, number and composition), a_k where ($k = 1...n$), vertical velocity, w , temperature, T , and pressure, p , such that it can be expressed as $N_a(a_1, ..., a_n, T, p, w)$. The average number of activated aerosols within a grid-box is denoted as $\bar{N}_a$.

UKCA-Activate requires the following fields from HadGEM3-A: pressure, temperature, specific humidity, saturated vapour pressure, air density and large-scale vertical velocity. Liquid cloud fraction by volume, and cloud liquid water content are also required to enable time-averages of in-cloud droplet number concentration. Boundary layer turbulent kinetic energy may also be required, depending on the parameterisation of sub-grid-scale vertical velocity required. This topic is explored in depth in Chapter 4.

The aerosol properties required for the activation scheme can be divided into prescribed definitions and dynamically-evolving properties simulated by GLOMAP-mode. UKCA-Activate requires the following definitions of the aerosol modes and components relating to the particular configuration of GLOMAP-mode that has been selected for a specific simulation:

- Number of modes present, I and logical definition of which modes are set.
- Number of chemical components possible in each mode, J , and the logical definition of which components are set in each mode.
- Molar mass of each possible component, M_{sij} in kg mol^{-1} .
- Mass density of components, ρ_{sij} in kg m^{-3} .
- Number of dissociating ions for soluble components, ϵ_{ij} (set to zero for insoluble components).
- Geometric standard deviation of each mode, σ_i (fixed values, listed in Table 3.1).

Secondly, aerosol activation depends on the following aerosol properties, calculated by GLOMAP-mode at each timestep:

- Number concentration of aerosol particles in each mode, N_i in m^{-3} . (GLOMAP-mode generates a dimensionless number-equivalent volume mixing ratio which is similar to a volume mixing ratio, but with ‘particles per molecule of air’ rather than ‘molecules per molecule of air’. In UKCA-Activate, this is multiplied by the number density of air molecules, to obtain the number density of aerosol particles, as required by the activation scheme.)

- Mass mixing ratio of each component present in each mode, x_{ij} in kg (aerosol) / kg (air).
- Dry geometric mean radius of each log-normal aerosol mode, $a_{m,i}$ in metres (calculated and stored by GLOMAP-mode as dry diameter).

At every grid-point, at every timestep, these properties are used within UKCA-Activate to calculate $\overline{B}_i$, the mean hygroscopicity coefficient averaged over all J components of each mode i , from Eq. (2.10) as described in Sec. 2.2.1. (Currently the osmotic coefficient is set to $\phi_{ij} = 1$ for all i and j , but composition dependence of this parameter could be incorporated if this information were available in UKCA.)

Note that, in practice, only the modes listed as soluble in Table 3.1 are involved, since although they may contain insoluble components (black carbon, organic carbon, and, in the future, mineral dust), this is accounted for in the calculation of the mean hygroscopicity coefficient. However, since the insoluble modes are defined to only contain mixtures of the insoluble components, they are not hygroscopic and cannot act as CCN.

Once $\overline{B}_i$ has been calculated, it is used in conjunction with the dynamically-varying geometric mean radius of each aerosol mode, $a_{m,i}$ and the non-aerosol-dependent Köhler A coefficient, Eq. (1.12), to calculate the critical supersaturation required to activate an aerosol with dry radius $a_{m,i}$, for each mode, using Eq. (2.11),

$$S_{m,i} = \frac{2}{\sqrt{\overline{B}_i}} \left(\frac{A}{3a_{m,i}} \right)^{3/2}.$$

The parameters η_i , $f_i(\ln \sigma_i)$ and $g_i(\ln \sigma_i)$ are calculated for each mode, from their definitions in Sec. 2.2.1, and then used with ζ and $S_{m,i}$ to calculate $S_{\max}$ from Eq. (2.13). The total number activated (summed over all I modes), N_a is then calculated using Eq. (1.26),

$$N_a = \frac{1}{2} \sum_{i=1}^I N_i [1 - \operatorname{erf}(z_i)],$$

where

$$z_i \equiv \frac{2 \ln(S_{m,i}/S_{\max})}{3\sqrt{2} \ln \sigma_i},$$

as discussed in Sec. 1.4.1.

Vertical velocity in the reference model

As mentioned previously, ARG calculates the number of aerosols that would activate (N_a) as a function of vertical velocity, w (both η_i and ζ are functions of w). However, observations show that within a typical grid-box-sized volume of atmosphere there can be a substantial degree of sub-grid-scale variability of vertical velocity. The question of how to account for such variability in a GCM is the focus of the next two chapters. In the meantime, in the reference configuration of UKCA-Activate used for the model evaluation in this chapter, a Gaussian probability density function, $P(w)$, is used to represent the likely distribution of vertical velocity within each grid-box at each time-step, with mean equal to the variable large-scale vertical velocity and standard deviation fixed at $\sigma_w = 0.4 \text{ m s}^{-1}$. (This configuration is referred to as **sigw0.4**.) The average number of aerosols that could activate at each grid-point is then calculated by taking the expected value,

$$\overline{N_a} = \frac{\int_0^\infty N_a(a_1, \dots, a_n, T, p, w) P(w) dw}{\int_0^\infty P(w) dw}. \quad (3.1)$$

Assumptions in calculation of grid-box mean cloud droplet number concentration

In UKCA-Activate, this average number of activated aerosols, $\overline{N_a}$, is used as a direct approximation of the number of cloud droplets in a grid-box, N_d . Since HadGEM-UKCA does not have a two-moment cloud scheme, the effect of other sources and sinks of cloud droplets (such as advection, mixing, evaporation, collision/coalescence, sedimentation and freezing) cannot be considered explicitly. Consequently, UKCA-Activate makes the simple assumption of local equilibrium between the number of aerosols activated at cloud-base and the number of droplets that exist at any one time step, and N_d is diagnosed directly from $\overline{N_a}$ (i.e. $N_d = \overline{N_a}$).

Parcel model simulations demonstrate that the supersaturation in a cloud is usually maximal just above cloud base, and that this is where most droplet activation occurs. For this reason, UKCA-Activate makes the further assumption that N_d throughout the vertical extent of a cloud is equal to the number of aerosols activated at cloud-base. This assumption extends over all vertically contiguous cloudy grid-boxes, which is consistent with the cloud overlap assumptions made in the radiation scheme that cloud in vertically adjacent grid-boxes overlaps maximally in its horizontal extent, within a column.

A criticism of this conceptual model relates to the scales over which the assumption is valid. While the interpretation may be valid on horizontal scales of tens of metres, similar to those used in parcel and column model studies, in a GCM the scales are of order hundreds of kilometres, and there may be no meaningful connection between cloud in one vertical layer and cloud in the layer above. It would therefore be interesting to perform a study to quantify the sensitivity of final results to this assumption.

3.3 Indirect aerosol effects from UKCA-Activate

Once this cloud droplet number concentration has been calculated by UKCA-Activate, it is used to model the indirect aerosol radiative effects in HadGEM-UKCA following the same method described by Jones et al. (2001). This is outlined briefly below.

3.3.1 Link between N_d and radiation

Cloud droplet number concentration is used in the calculation of liquid cloud droplet effective radius (r_e), as described in Sec. 1.5.2. This allows the first indirect aerosol effect, the cloud albedo effect, to be modelled. In the Edwards–Slingo radiation code in HadGEM3, this effective radius is parameterised following Martin et al. (1994), i.e.

$$r_e = \left(\frac{3}{4\pi} \frac{\rho_{\text{air}} q_c}{\rho_w k N_d} \right)^{1/3}, \quad (3.2)$$

where q_c is the cloud liquid water content (kg kg^{-1}) and ρ_{air} and ρ_w the densities of air and water. The value of k is set to 0.67 over land and 0.8 over ocean (Martin et al., 1994).

In fact, in Eq. (3.2), $r_e \propto N_d^{-1/3}$. In UKCA-Activate, N_d is defined as the expected value of the number of activated aerosols over a pdf of vertical velocity. Since $\overline{(N_a^{-1/3})} \neq (\overline{N_a})^{-1/3}$, the expected value of $[N_a(w)]^{-1/3}$ is also calculated by UKCA-Activate:

$$\overline{N_a^{-1/3}} = \frac{\int_0^\infty [N_a(w)]^{-1/3} P(w) dw}{\int_0^\infty P(w) dw}, \quad (3.3)$$

and this is what is used in the effective radius calculation (see Morales and Nenes (2010)).

3.3.2 Link between N_d and precipitation rate

In the large-scale precipitation scheme, the rate of increase of rain water by autoconversion of cloud droplets to precipitation, R_{auto} , is parameterised as

$$R_{\text{auto}} = \frac{0.104gE_c\rho_{\text{air}}^{\frac{4}{3}}q_c^{\frac{7}{3}}}{\mu\rho_w^{\frac{1}{3}}N_d^{\frac{1}{3}}}, \quad (3.4)$$

where g is the acceleration due to gravity and μ is the dynamic viscosity of air. E_c is the collision/collection of cloud droplets (assumed to be 0.55 by Tripoli and Cotton (1980)). Within the scheme, autoconversion occurs when the liquid water content is above a certain threshold, and is also dependent on N_d (and hence aerosol). Autoconversion is allowed to proceed when the concentration of cloud droplets with radius greater than $20\text{ }\mu\text{m}$ exceeds $1 \times 10^3\text{ m}^{-3}$ (Rogers and Yau, 1989), found by assuming a Khrgian–Mazin modified gamma cloud droplet size distribution (e.g. Pruppacher and Klett (1997)).

This dependence of precipitation rate on cloud droplet number concentration, derived from UKCA aerosol properties, allows the effect of aerosol on precipitation efficiency, leading to the cloud lifetime effect, to be modelled.

3.4 Reference model configuration

All the model simulations used in this thesis have the same fundamental set-up. HadGEM-UKCA was run for a year (preceded by an additional three-month spin-up period), in atmosphere-only mode, with sea-surface temperatures and sea-ice extent prescribed as seasonally varying fields. Meteorology for all runs was nudged to ECMWF ERA-40 reanalysis data, as described by Telford et al. (2008).

Present-day aerosol emissions (used in all simulations in Chaps. 3, 4 and 5) are taken from the Coupled Model Intercomparison Project Phase 5 (CMIP5) emissions dataset (Lamarque et al., 2010) for the year 2000, provided to support modelling activities for the IPCC fifth assessment report (AR5). For biomass burning emissions, grass-fire emissions are added to the surface layer, and forest fires are assumed to be equally distributed between model levels two and twelve (approximately from 50 m up to 3 km). Dimethylsulfide (DMS) emissions are estimated from the interactive scheme in HadGEM3, and used to drive UKCA.

3.5 Comparison between modelled and observed CCN

In order to evaluate the ability of UKCA to produce cloud-relevant aerosol, online diagnostics were added to calculate the number of CCN that could form at each of 38 distinct values of supersaturation, S , corresponding to the most common values of S in a database of measurements documented by Spracklen et al. (2011). These measurements were compiled from 55 separate studies in the peer-reviewed literature, and include 278 observations made at 80 locations across the globe. Measurements were made by independent research groups over the years 1971–2009, using a variety of instruments, with S ranging from 0.02% to 1.5%.

In the model, CCN concentrations are calculated from aerosol in the lowest model level only, since the majority of observations in the database are from surface sites (including several from ship-borne instruments). Of the few measurements made above the surface, only those made within the lowest two kilometres of the atmosphere are included.

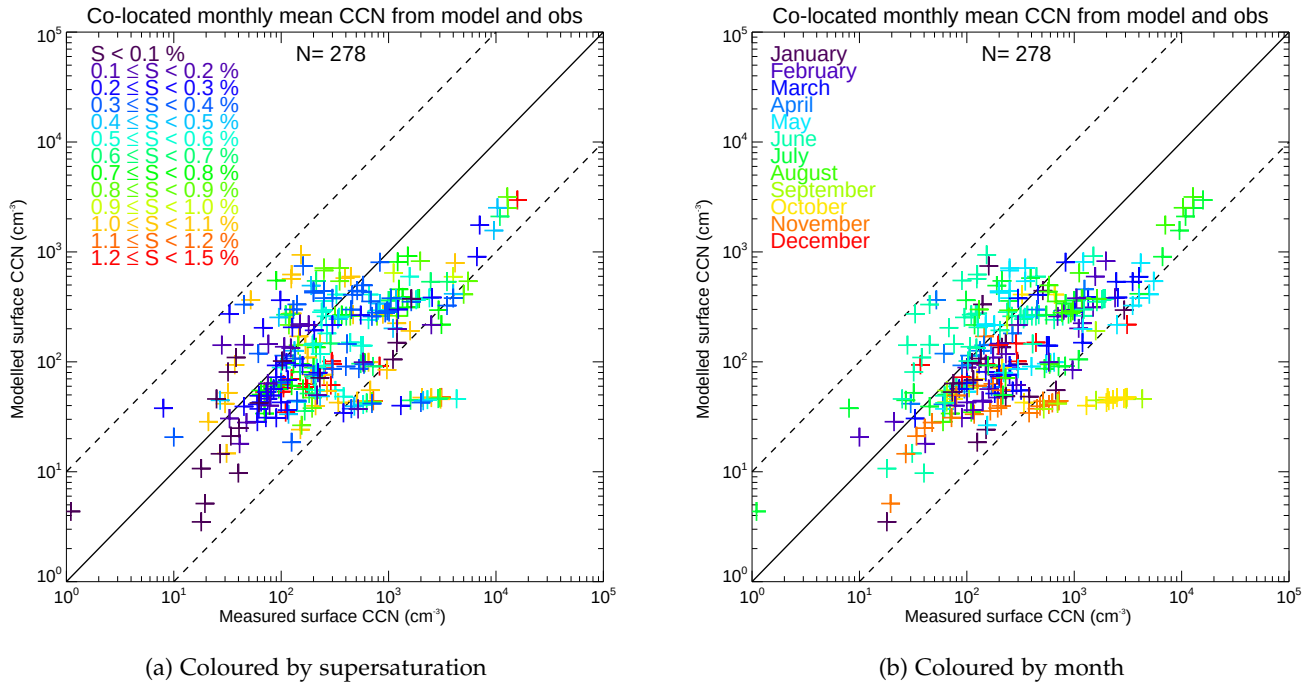


Figure 3.1: CCN measurements from 55 separate studies, plotted against the monthly mean modelled surface concentration of CCN at the same supersaturation and location. Points are coloured by (a) supersaturation and (b) month.

Following the method set out by Abdul-Razzak and Ghan (2000) for multi-modal aerosol distributions, the dry radius $a_{\text{fix},i}$ of the smallest particle that could activate in each mode, i , at a specified supersaturation, S_{fix} , is calculated using

$$a_{\text{fix},i} = \frac{A}{3} \left(\frac{4}{\overline{B}_i S_{\text{fix}}^2} \right)^{\frac{1}{3}}, \quad (3.5)$$

where A is the Köhler coefficient of the curvature effect (independent of aerosol properties) defined in Eq. (1.12), and $\overline{B}_i$ is the mean hygroscopicity coefficient averaged over all J components of each mode i , defined in Eq. (2.10).

The number concentration of CCN at $S = S_{\text{fix}}$ is then calculated by

$$N(S_{\text{fix}}) = \frac{1}{2} \sum_{i=1}^I N_i \left[1 - \text{erf} \left(\frac{\ln(a_{\text{fix},i}/a_{\text{m},i})}{\sqrt{2} \ln \sigma_i} \right) \right], \quad (3.6)$$

from Eq. (1.26), where N_i is the number concentration of aerosols in mode i and $a_{\text{m},i}$ is the geometric mean radius of mode i (both dynamically-varying properties of the aerosol distribution). σ_i is the fixed geometric standard deviation of mode i , listed in Table 3.1.

Fig. 3.1 shows scatter plots of measured versus modelled CCN number concentrations, matched by month, location and supersaturation. Measurements taken during observation periods lasting less than a month are plotted against the monthly mean modelled CCN number concentration at the closest value of S , at the nearest horizontal grid-point. Measurements from longer observation periods are plotted against the average of the modelled CCN over that period. Points are coloured by supersaturation or month.

90% of points are within a factor of ten of the 1:1 line and the Pearson product-moment correlation coefficient between the logarithms of the measured and modelled CCN concentrations is $r = 0.591$. The visible group of outliers at measured CCN of order $1 \times 10^3 \text{ cm}^{-3}$ corresponds to two studies in the Amazon region in late September and early October, and the model clearly underestimates the number of CCN. This could be due to an underestimate of the number of CCN created by biomass burning, or a systematic error due to the lack of secondary organic aerosol within this configuration of UKCA.

The studies in the database include measurements made at 50 different values of supersaturation, with S ranging from 0.02% to 1.5%, although some values of S were far more common than others. As stated previously, modelled CCN are calculated at the 38 most common values of S within this group, which encompasses 245 out of 278 measurements. These 245 measurements are plotted against modelled CCN at the exact same supersaturation, in Fig. 3.1. The remaining 33 measurements are plotted against modelled CCN at the closest value of S .

Four examples of global maps of monthly mean modelled CCN with overlaid measurements are shown in Fig. 3.2. The first three maps display how the modelled CCN concentration varies with increasing supersaturation, and how this varies from region to region. In some regions, such as off the west coast of Chile, the number of CCN does not increase with increasing S , because the number of soluble aerosols produced by the model at this location is low, and the high fraction of activation at low S cannot increase much more with further increase to S . However, in many other regions, particularly over land, the abundance of soluble aerosol particles due to industry and biomass-burning means that significant increases in the CCN concentration are possible as S increases.

Fields of modelled CCN reflect some of the trends and spatial gradients seen between the overlaid measurements. For example, the increasing number of CCN at $S = 1\%$ going from the west to the east coasts of North America as seen in the three measurements is also reflected by the spatial gradient in the model. Off the coast of California, the increase in the measured CCN at $S = 0.4\%$ compared to $S = 0.2\%$ is also matched in the model. Interestingly, while there is an observed increase in CCN from 0.2% to 0.4%, there is less of an increase in this region in both the measurements and model from 0.4% to 1.0%, indicating that the fraction of activation is saturated in both model and observations. However, in other regions it is clear that although the model captures the general trends in the observations, the magnitude is often underestimated. This can be seen clearly in the fourth map of CCN at $S = 0.38\%$ in May. Here measurements from a ship cruise in the north Atlantic are higher in number than the modelled values beneath. Although the model does manage to capture the elevated level of CCN over eastern North America, and the gradient to the less elevated but still heightened levels over the eastern north Atlantic, the magnitude is lower than that measured.

The underprediction of CCN by UKCA is currently under active investigation, and several possible contributing factors are suggested here. First, in the version of UKCA used for the simulations in this thesis, the emission, deposition and boundary layer mixing of aerosols are treated in an operator split fashion. This means that these processes occur sequentially (and in this order), each time UKCA is called. While this may be a legitimate strategy in the case that surface fluxes are small, a problem arises when surface fluxes are large, such as with the emission of sea-salt at the ocean surface. In this case, large amounts of sea-salt are emitted into the lowest model level but, before any mixing can occur, dry deposition then acts immediately to remove much of this freshly-emitted aerosol. In the next release of UKCA, this operator splitting will be removed, to allow these three processes to be treated simultaneously. This should lead to more realistic CCN concentrations.

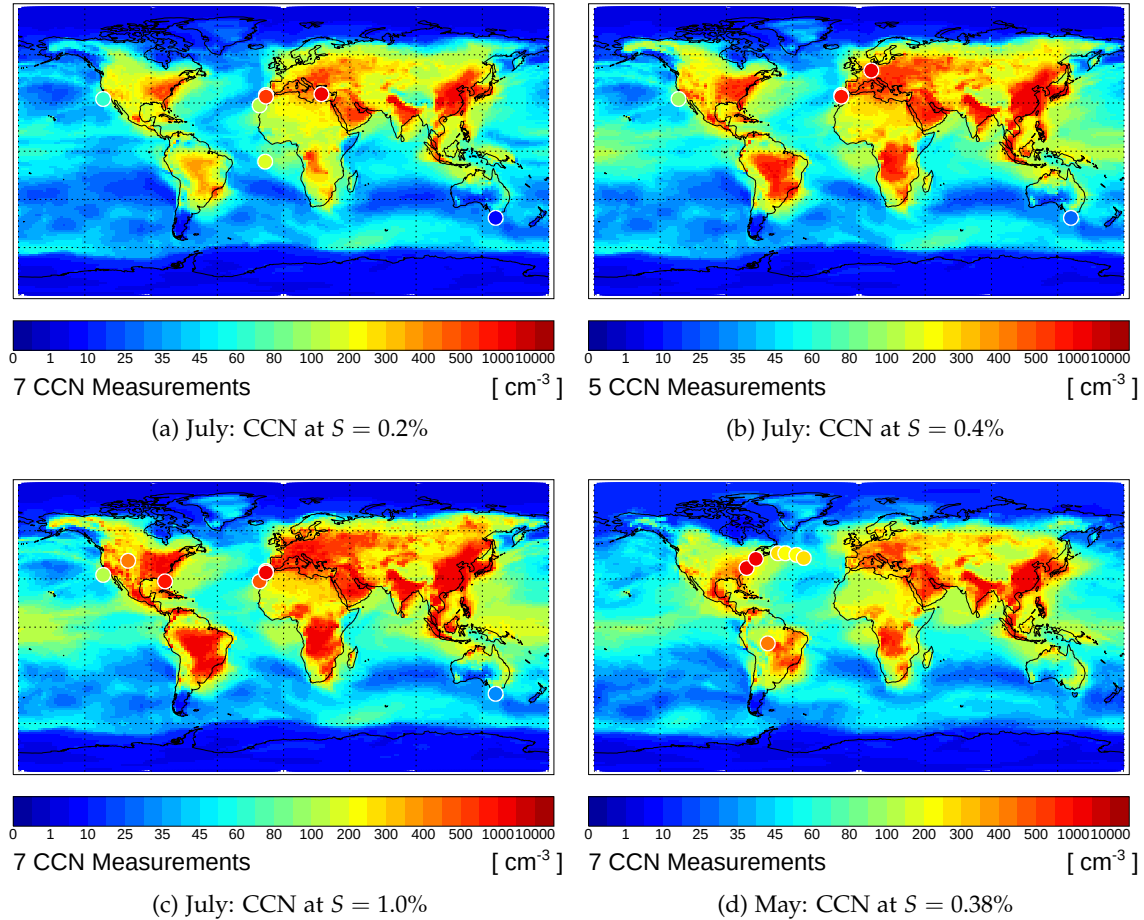


Figure 3.2: Maps of monthly mean modelled surface CCN for selected combinations of fixed supersaturation and month, noted in sub-captions. Measurements made at the same supersaturation in the same month are over-plotted in circles edged in white. The number of measurements in each graph is noted underneath. (In some cases, measurements made in close proximity to one another overlap.)

Another factor which could contribute to the underprediction of CCN in stratocumulus regions is that the Hadley Centre model produces too much drizzle in these regions, in both numerical weather prediction (NWP) and climate resolutions (Bodas-Salcedo et al., 2011; Wilkinson et al., 2012). Excessive drizzle production leads to an overestimation of the amount of aerosol that is removed by wet deposition and scavenging in UKCA, and hence an underprediction of the CCN concentration.

Recent work by Boutle and Abel (2012) has investigated the simulation of stratocumulus cloud and drizzle in the NWP configuration of the Hadley Centre model. This study used high-resolution simulations of the south east Pacific stratocumulus-topped boundary layer and compared these to observations made during the VOCALS-REx campaign (Wood et al., 2011). They found that implementing a simple change to the rain drop size distribution (Abel and Boutle, 2012) and changing the autoconversion parameterisation from Tripoli and Cotton (1980), as used in HadGEM-UKCA, to Khairoutdinov and Kogan (2000) improved the simulation of both cloud and precipitation compared with observations. Further work will now be required to assess whether these updates are appropriate for regimes other than

stratocumulus, and hence for adoption in the global climate configuration of the model.

A further factor which contributes to the underprediction of CCN by UKCA is also related to rainfall. In the current configuration of UKCA, the process by which below-cloud aerosol is scavenged, and hence removed from the atmosphere by precipitation, ignores the evaporation of raindrops. This means that any aerosol contained within an evaporating droplet is unrealistically deposited at the surface, rather than staying aloft, thus depleting potential CCN concentrations. This exacerbates the effect of the excessive amounts of drizzle mentioned previously, leading to even greater reductions in CCN concentrations in stratocumulus regions.

In summary, the modelled CCN have been evaluated against a database of over 250 measurements, at a variety of locations, seasons, and at a range of supersaturations. Although modelled CCN concentrations are biased low compared to measurements, in some regions, they are found to be within a factor of ten of the measured values in 90% of cases, and the low bias is under active investigation.

3.6 Comparison of model with satellite data

This section compares annual mean model output with data from three Earth-observing satellite datasets. Aqua-MODIS data from two separate datasets are used: firstly, the official MODIS Science Team Atmosphere Collection 5 level 3 daily $1^\circ \times 1^\circ$ cloud and aerosol dataset (MYD08_D3, 1/12/2002 to 30/11/2008); secondly, the CERES Single Scanner Footprint (SSF) dataset, which comprises the CERES team cloud data, combined with the MODIS Science Team Collection 4 aerosol data, gridded to $2.5^\circ \times 2.5^\circ$ (1/12/2002 to 30/11/2005). Thirdly, the AATSR GlobAEROSOL dataset is combined with the AATSR ORAC cloud dataset, gridded to $1^\circ \times 1^\circ$ (1/12/2002 to 30/11/2006). Annual means are calculated from level 3 daily data, over the periods defined. (MODIS level 3 data were provided by Benjamin Grandey; CERES SSF level 3 data were produced by Johannes Quaas, see Quaas et al. (2009); AATSR level 3 data were produced by Christopher Arnold.)

Fig. 3.3 shows the annual mean aerosol optical depth (AOD) at $\lambda = 550$ nm as retrieved in each of these satellite datasets, alongside that from model configuration sigw0.4 (see Table 4.2). In these figures, and in all others marked ‘SATSAMP’, model output is sampled at runtime at the Aqua-MODIS satellite overpass time of $13:30 \pm 30$ min.

This set of figures, and the similar sets which follow, comprises the annual mean retrieved quantity from three different satellite datasets as well as the model reference case, sigw0.4. For each set of figures, the focus of this section will be a comparison between the model output and the group of satellite retrieved properties, rather than a detailed analysis of the differences between the satellite datasets (given in e.g. Grandey (2011)). Possible reasons for differences between the satellite datasets will only be given where they are of direct relevance to the model comparison, although it is noted that the comparison between MODIS Collection 5 and the CERES SSF datasets is not necessarily a fair comparison between MODIS Collections 4 and 5, because the quality control may differ between the production of the two level 3 datasets.

In some cases (e.g. Fig. 3.6), it is clear that inter-satellite differences are just as great—if not greater—than the differences from the model. These cases serve to illustrate that despite ongoing work to improve satellite retrievals, in some cases these satellite datasets can only provide a weak observational constraint on the model predictions, and further work is still required on the development of retrieval algorithms for aerosol and cloud properties.

For AOD, the three satellite datasets show broad agreement, particularly over ocean.

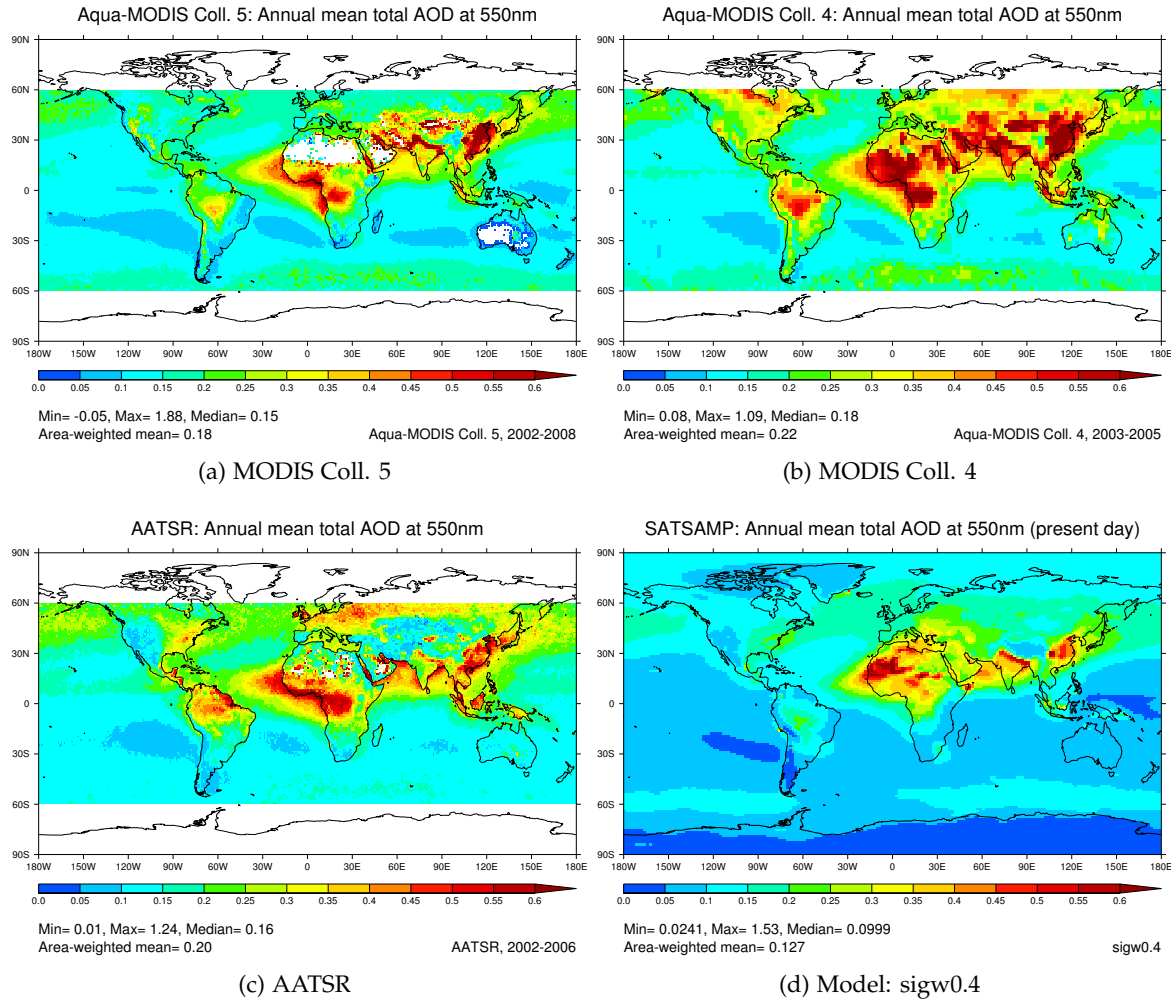


Figure 3.3: Annual mean aerosol optical depth at $\lambda = 550$ nm from (a) Aqua-MODIS Collection 5 (b) Aqua-MODIS Collection 4 AOD with CERES SSF cloud data (c) AATSR GlobAEROSOL AOD with ORAC cloud and (d) model configuration sigw0.4, sampled at Aqua-MODIS overpass time (13:30 local time).

One difference is in the Southern Ocean, where AATSR reports significantly lower AOD than the two MODIS retrievals. This is likely due to the fact that the GlobAEROSOL retrieval has a wind speed dependent sea-surface brightness correction (Sayer et al., 2011). Missing data over the Sahara desert and differences in retrievals over Australia are attributable to the difficulty in performing satellite retrievals of aerosols over bright surfaces (Remer et al., 2005). AATSR exhibits particularly strong land/sea contrasts in some regions, which are not seen in the AOD retrieved by MODIS (e.g. along the north-eastern coast of South America; the British Isles; Borneo, Sumatra and the Malay Peninsula; the Red Sea, Persian Gulf, Arabian Gulf, and the Bay of Bengal).

In terms of aerosol retrievals, a useful intercomparison between AATSR and MODIS (and several other satellite products), is provided by the GlobAEROSOL product validation and intercomparison report (Poulsen et al., 2009). The report also includes an independent evaluation of these satellite datasets against the Aerosol Robotic Network (AERONET, Holben et al. (2001), a global network of ground-based sun photometers providing long-

term records of aerosol optical properties.

Although the datasets exhibit some strong agreement, the report emphasises how disagreements are variable both spatially and temporally. Results of comparisons are presented separately by region as maps and time series. In general, the time series show that aerosol have strong seasonal cycles in most regions, and that the agreement between satellite datasets often differs depending on the time of year. This makes regionally- and seasonally-specific findings hard to summarise, but there are a few general results. For instance, over land, MODIS AOD generally compares better against AERONET. (The deficiencies of AATSR over land are particularly evident in the contrasts visible along the coastlines listed previously.) The report states that AATSR AOD over land could be improved if only retrievals in which all channels are present were used. On the other hand, AATSR is found to capture the seasonal cycles in particle size more accurately than MODIS, when compared to AERONET. General problems for the retrieval of AOD from all satellites stem from differences in cloud flagging (i.e. aerosol being mistaken for cloud or *vice versa*), problems with modelling surface reflectance over both land and sea, and finally due to difficulty in the identification of different aerosol species. These are all challenges that current developments are trying to address.

The model captures the large-scale features of the satellite datasets, such as the dust emitted from the Sahara, and the heightened anthropogenic aerosol burden over the Eurasian land mass, in particular India and China. Over ocean, the model does well in the northern hemisphere, for example, in capturing the increased aerosol in the Asian outflow region over the Pacific. Increased sea-salt burden in the high winds over the extra-tropical Southern Oceans is also evident in the model, albeit not as strongly as in the MODIS retrievals, but, as stated previously, AATSR is probably more reliable than MODIS in this region, due to the sea surface brightness correction applied in the retrieval. Other than this, modelled AOD is slightly lower than that retrieved by MODIS over most of the oceans of the southern hemisphere, although this difference is intensified by the non-linear colour scale. The model also shows lower AOD in the stratocumulus regions off the western coast of South America, as compared with all three satellite datasets (this corresponds with the low values of CCN concentration in this region of the model).

Over land, the model generates lower AOD than the satellites in northern South America and sub-Saharan Africa, although retrievals over these land regions are not necessarily reliable. Modelled AOD over North America is also significantly lower than that retrieved from satellites, although the satellites do not agree on the spatial distribution of aerosol in this region. Again, satellite retrievals over land are less reliable than over ocean (Platnick et al., 2003; Remer et al., 2005).

3.6.1 Cloud droplet number concentration

Cloud droplet number concentration is not a quantity directly retrieved by satellites but, as discussed by Grandey (2011), it may be estimated at the cloud-top by the adiabatic approximation,

$$N_d = \gamma \tau_c^{\frac{1}{2}} r_e^{-\frac{5}{2}}, \quad (3.7)$$

where τ_c is the retrieved cloud optical depth² (Platnick et al., 2003); r_e is the two-dimensional retrieved liquid cloud droplet effective radius at the cloud-top; and $\gamma = 1.37 \times 10^{-5} \text{ m}^{-\frac{1}{2}}$ (Brenguier et al., 2000b; Quaas et al., 2006) or $\gamma = 1.125 \times 10^{-5} \text{ m}^{-\frac{1}{2}}$ (Bennartz, 2007; George

²cloud optical depth is the total extinction at a given wavelength due to cloud water (liquid and/or frozen) in a column

and Wood, 2010)), depending on assumptions regarding atmospheric conditions³. Again, following Grandey (2011), the Quaas et al. (2006) value of $\gamma = 1.37 \times 10^{-5} \text{ m}^{-\frac{1}{2}}$ is used, as well as the Quaas et al. method of excluding clouds with $\tau_c < 4$ and $r_e < 4 \mu\text{m}$ when calculating N_d . The Quaas et al. (2006) satellite-derived N_d are calculated from the MODIS retrievals of τ_c and r_e only for pixels where the retrieval is most reliable, which has been shown to be in the interval $4 \leq r_e \leq 30 \mu\text{m}$ and $4 \leq \tau_c \leq 70$ (Nakajima and King, 1990). As discussed by Grandey (2011), removing clouds with $\tau_c < 4$ implicitly removes clouds with low liquid water path in the calculation of N_d (Eq. (3.7)). For clouds with a typical $r_e = 10 \mu\text{m}$ this corresponds to excluding clouds with liquid water path (column-integrated cloud liquid water content) less than 27 g m^{-2} .

The physical assumptions inherent in Eq. (3.7) make it most suitable for non-precipitating stratocumulus cloud fields with little entrainment and it has been shown to be in good agreement with aircraft measurements of N_d in stratocumulus clouds in the south-east Pacific with cloud fractions greater than 0.8 (Bretherton et al., 2010). Converting from satellite-retrieved cloud-top r_e to give a two-dimensional field of N_d at the cloud-top in this way, using Eq. (3.7), can be shown to be effectively the inverse of the model calculation of r_e from N_d in Eq. (3.2), which uses the same the adiabatic approximation (but different constants of proportionality).

Comparison between model and satellites

Fig. 3.4 shows the annual mean liquid cloud droplet number concentration derived from the three satellite datasets described in Sec. 3.6 using Eq. (3.7). All three satellites show a marked contrast between values of N_d over land and ocean, although the absolute values of N_d from AATSR are significantly higher than those from both MODIS retrievals. These high values of N_d are a direct result of the smaller droplet effective radii retrieved by AATSR, which will be seen in Fig. 3.5. [Differences between the retrieved r_e are amplified by the power law relationship in Eq. (3.7), resulting in even greater differences in N_d .]

The main reason for the differences between the liquid cloud properties retrieved by AATSR, MODIS Science Team and MODIS CERES SSF is due to the choice of wavelengths used in the retrievals, which leads the instruments to sample different penetration depths within the clouds in some cases. These differences are discussed in detail by Sayer et al. (2011) but, essentially, AATSR uses the $1.6 \mu\text{m}$ channel, whereas MODIS Science Team uses $2.1 \mu\text{m}$ and MODIS CERES $3.7 \mu\text{m}$. While these differences hardly matter in clouds with a constant vertical profile of droplet size, non-precipitating clouds typically show an increase in particle size with height (see Platnick et al. 2000). Since the absorption coefficient of water increases with wavelength, the shorter wavelength of the AATSR penetrates deeper into such clouds. This largely explains why r_e from AATSR appear so much smaller than from MODIS, and hence why AATSR N_d appear higher.

In an attempt to compare like with like, modelled cloud properties are sampled at the liquid cloud top. This is defined as the first vertical level in a column to contain liquid cloud water, starting at the top of the atmosphere in each column and working downwards (there is no masking caused by ice clouds). This gives a two-dimensional sample from the three-dimensional cloud field (maximum/random overlap of clouds is assumed in the

³It can be shown that γ in Eq. (3.7) is proportional to β^2 , where β represents the dispersion of the cloud droplet spectrum (Eq. (1.38)). The value of β is obtained empirically, and is highly variable (Martin et al., 1994; Liu and Daum, 2002; Rotstajn and Liu, 2003). The different choices of gamma used in different retrievals of N_d reflect the variable nature of cloud droplet spectral dispersion.

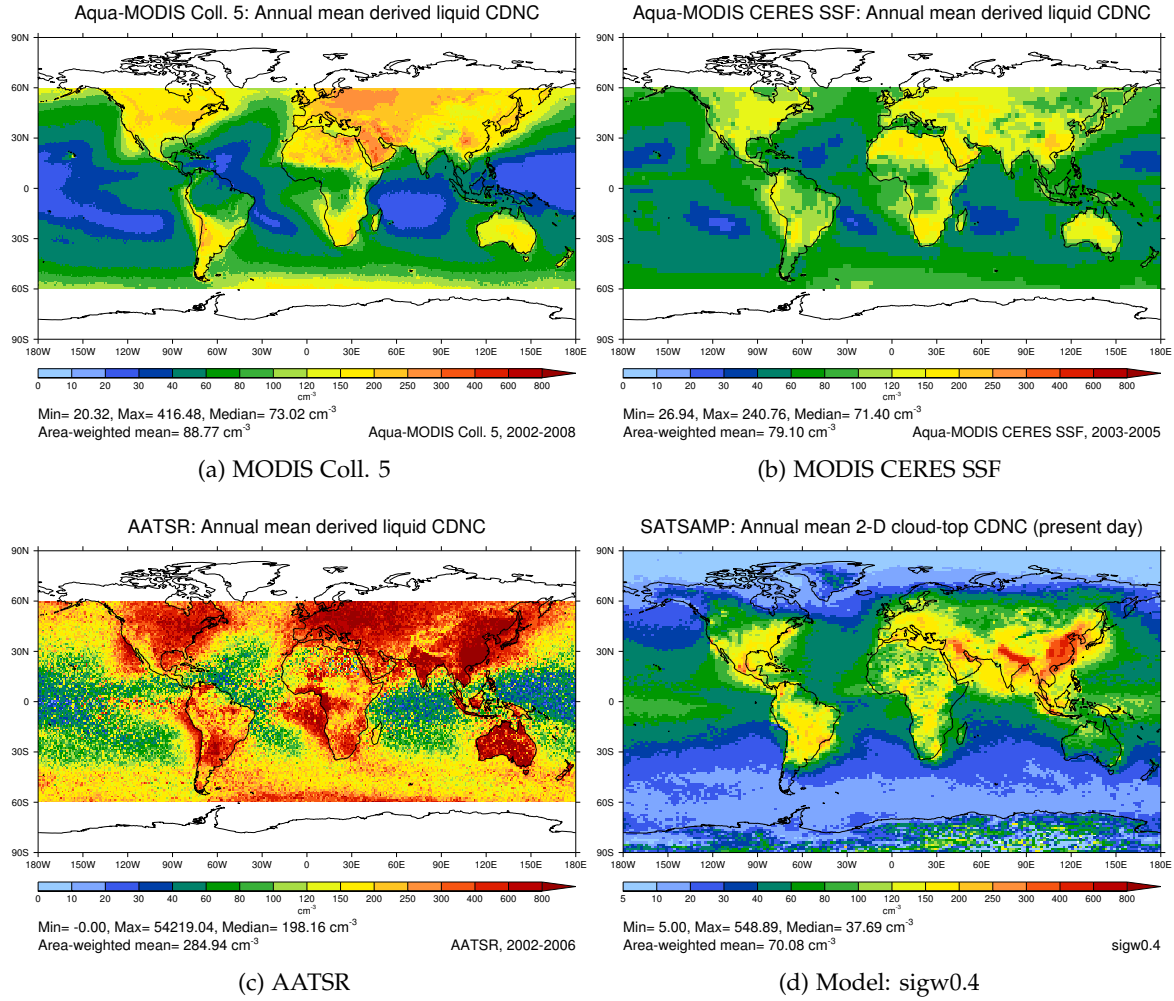


Figure 3.4: Annual mean liquid cloud droplet number concentration at cloud-top derived from (a) Aqua-MODIS Coll. 5, (b) Aqua-MODIS CERES SSF cloud data, (c) AATSR, and (d) model configuration sigw0.4, sampled at cloud-top at Aqua-MODIS overpass time (13:30 local time). Note non-linear colour scale.

model), which can be compared with the retrieved liquid cloud properties retrieved from satellite data.

In general, modelled N_d shows a similar land/sea contrast to the satellites, although absolute values tend to be slightly lower. From a modelling perspective, lower values of modelled N_d can be attributed either to a lack of suitable CCN, or a deficit of supersaturation, potentially due to weak updraught velocities (or in some cases due to the presence of giant CCN). Later work in this thesis will investigate whether changes in N_d can be attributed to one mechanism or the other, but the inter-linked nature of the two makes this a non-trivial problem.

Notwithstanding the differences in absolute values of retrieved N_d mentioned previously, there are several notable differences between the model and all three satellite datasets. First, in the persistent stratocumulus regions, all three satellites show a strong relative increase in N_d in the stratocumulus regions off the west coasts of California, South America, and southern Africa; however, in the model, the latter two regions are barely visible, due to the

low values, and the increase in N_d over the north-east Pacific is smaller in magnitude and geographical extent in the model than in the observed values.

Second, modelled N_d in the extra-tropical southern oceans is at least 40 cm^{-3} lower than the values measured by MODIS and MODIS CERES SSF, and up to 400 cm^{-3} lower than the most southerly values recorded by AATSR. This is despite the increased modelled AOD over the Southern Ocean mentioned in the previous section.

As well as these deficiencies, there are other contrasting regions where the model over-predicts N_d compared to MODIS. For instance, the model generates much higher N_d over the strongly polluted region of China (corresponding to the increased model AOD in this region, shown previously in Fig. 3.3), and is closer to the values of N_d retrieved by AATSR. Modelled values of N_d are also higher than MODIS over the whole Indian sub-continent, and particularly elevated over the Ganges plain. (It is interesting to compare this feature in modelled N_d with the modelled AOD in Fig. 3.3. It is perhaps made more prominent by the sharp decrease in aerosol over the Tibetan plateau.) The surrounding oceans also see elevated N_d in the model, over the Arabian Sea and Bay of Bengal. Less marked increases are also visible in the Amazon, and over sub-Saharan Africa.

Overall, modelled cloud droplet number concentration compares reasonably well with both MODIS retrievals in terms of magnitude, and with all three satellites in terms of spatial distribution. Potentially important deficiencies may exist in several of the persistent marine stratocumulus regions.

3.6.2 Cloud droplet effective radius

In some ways, cloud-top liquid cloud droplet effective radius (r_e) is a more meaningful property than N_d to consider when inter-comparing satellite datasets, since this is what is directly retrieved, and does not rely on the adiabatic assumption in Eq. (3.7), needed to convert r_e to N_d . Having said that, since the same assumption is used in reverse to obtain modelled r_e from N_d , the adiabatic assumption is inescapable when trying to make inter-comparisons between satellite and model data. While N_d is perhaps a more intuitive cloud property to consider, it is also useful to consider the effective radius of the droplets (and hence the total surface area of N_d droplets) which controls how a cloud of a particular liquid water content interacts with incident radiation. (This is, of course, also one of the reasons why effective radius can be deduced from the radiances measured by a radiometer.)

Annual mean values for the three satellite datasets and reference model are shown in Fig. 3.5. AATSR retrieves much smaller effective radii over ocean than MODIS, although the spatial distribution is similar, but offset. This is due to the choice of wavelength used by the different instruments, as described previously. However, the land/sea contrast is stronger in the MODIS data, particularly in the MODIS science team retrieval (as previously seen in the values of N_d in Fig. 3.4).

Modelled effective radii are generally larger than AATSR r_e , but compare favourably with those from MODIS, particularly MODIS CERES SSF. As expected, the spatial distribution of features is roughly the inverse of the modelled cloud-top N_d , due to the relationship between the two ($r_e \propto N_d^{-1/3}$). This leads to low values of effective radii in regions of high N_d (e.g. China). The strongest short-wave cloud forcing will occur in regions where the frequency of occurrence of clouds is high, the cloud fraction is high, the liquid water path is high, and the effective radii of the droplets is small, which highlights the particular importance of accurate modelling of N_d (and hence r_e) in the persistent stratocumulus regions. The model does relatively well in the north-east Pacific stratocumulus region, where modelled r_e are of similar size to those observed by all three satellites. However, in the

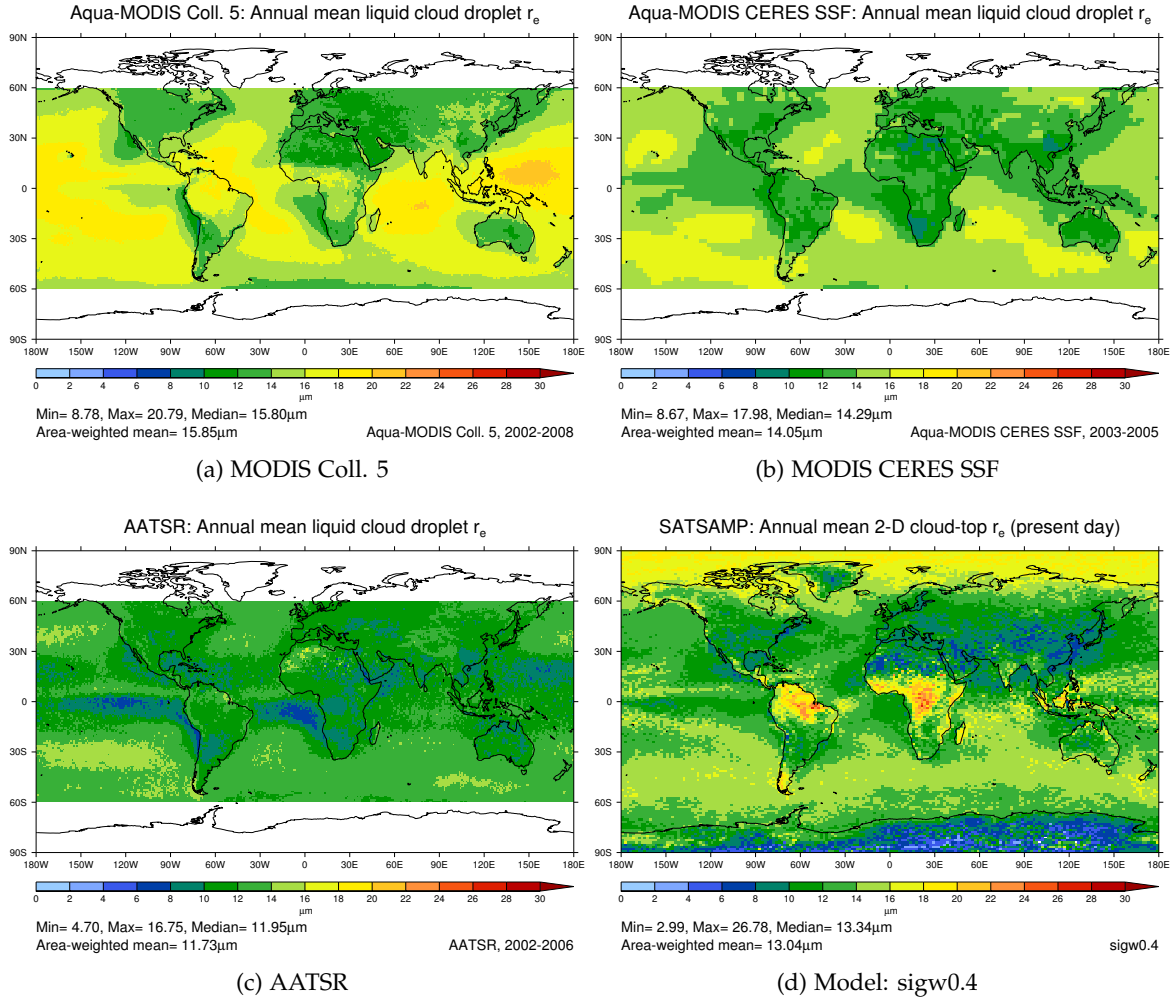


Figure 3.5: Annual mean liquid cloud droplet effective radius from (a) Aqua-MODIS Coll. 5, (b) Aqua-MODIS Coll. 4 aerosol data with CERES SSF cloud data (c) AATSR ORAC cloud dataset and (d) model configuration sigw0.4, sampled at cloud-top at Aqua-MODIS overpass time (13:30 local time).

southern hemisphere oceans, the r_e in the stratocumulus regions in the satellite retrievals are smaller than in the model. This is true to a greater extent off the west of Chile, and to a lesser extent off the west coast of southern Africa. As described above, this could be due to a lack of modelled CCN in these regions.

The large modelled r_e in the tropical land regions (e.g. Indonesia, sub-Saharan Africa and central America) occur partly because of the high liquid water path in these regions (this will be shown next, in Fig. 3.6). Interestingly, these regions of the model exhibit a strong diurnal cycle in liquid water path (LWP), which is maximal during the day, roughly coincident with the sampling of diagnostics at the satellite overpass time. N_d does not exhibit such a strong daily cycle in these regions, but since r_e is also dependent on LWP where N_d is not, the effect is noticeable in the field of r_e . These regions of larger r_e are far less intense in the annual mean over all time steps (not shown) than when the field is restricted to being sampled at 13:30. However, the meteorology in these regions is dominated by convection, and the activation of aerosol in strong convective updraughts is

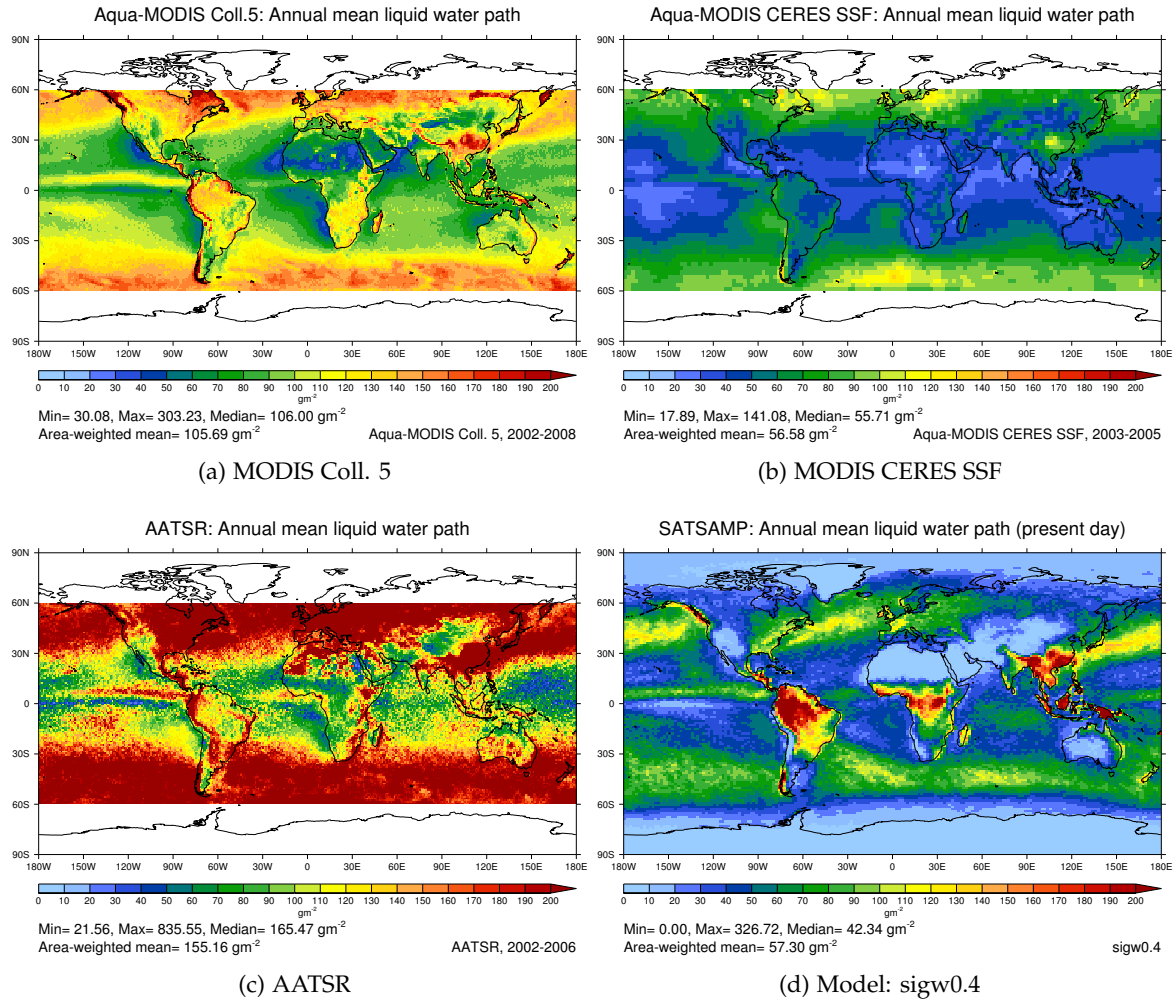


Figure 3.6: Annual mean liquid water path from (a) Aqua-MODIS Coll. 5, (b) Aqua-MODIS Coll. 4 aerosol data with CERES SSF cloud data (c) AATSR ORAC cloud dataset, and (d) model configuration sigw0.4, sampled at Aqua-MODIS overpass time (13:30 local time).

currently beyond the scope of UKCA-Activate.

3.6.3 Liquid water path

Annual mean liquid water path (column-integrated cloud liquid water content) from the three satellites and the reference model is shown in Fig. 3.6. Spatial patterns are similar between all three satellite products, but there is no consensus on the magnitude of this quantity. This is partly due to the way that multi-layer clouds are treated by the different instruments. For instance, the MODIS Science Team retrieval ignores some optically thin clouds that are not ignored by the MODIS CERES retrieval, which explains the bias between the two (Minnis et al., 2011). Similarly, the AATSR cloud flag is known to miss some thin clouds, particularly over the tropical oceans and yet, conversely, in the storm tracks, multi-layer clouds are often mistakenly attributed as liquid rather than ice clouds. In general, microwave-derived retrievals are thought to be more informative than those derived from the visible part of the spectrum for LWP over oceans (Sayer et al., 2011).

The model reflects the latitudinal variation of liquid water path seen in the satellite datasets, over ocean (i.e. low in the tropics, increasing towards the mid-latitudes) but over land the modelled liquid water path tends to be lower than that observed, except in the tropical regions mentioned previously. Modelled liquid water path in these regions also exhibits a strong diurnal cycle, as described previously. One major difference between the model and retrieved LWP is that all three satellites show a relative heightened region of LWP in the eastern subtropical South Pacific, which is only weakly reproduced in the model. It could be that the drizzle rate is too high in this stratocumulus region of the model, which decreases the cloud liquid water content.

3.7 Sensitivity of cloud properties to aerosol optical depth

One method of making quantitative estimates of aerosol–cloud relationships in satellite or model datasets is to calculate the so-called ‘sensitivity’ of a cloud property, ϕ , to aerosol optical depth, τ , where this sensitivity, b , is defined (following Quaas et al. (2008)) as

$$b_\phi = \frac{d \ln \phi}{d \ln \tau}. \quad (3.8)$$

Here, and throughout, aerosol optical depth at $\lambda = 550 \text{ nm}$ is used. As discussed in the previous section, three commonly retrieved cloud properties are of particular relevance to the indirect aerosol effects. These are the liquid cloud droplet number concentration ($\phi = N_d$), liquid cloud droplet effective radius ($\phi = r_e$), and cloud liquid water path ($\phi = W$). Sensitivities of each of these properties have previously been calculated by Grandey (2011), for each of the satellite datasets discussed in the previous section. This analysis has been repeated with appropriately sampled data from the HadGEM-UKCA reference model (in configuration sigw0.4, as used in the rest of the chapter, sampled at the satellite overpass time, with cloud properties sampled at liquid cloud-top), using the grid-method proposed by Grandey and Stier (2010). Figures from Grandey (2011) have been reproduced in this thesis for comparison with the model output, courtesy of the author. In this section, a brief overview of this analysis method is given, although the reader is referred to the original document for a full description and thorough discussion of the implicit assumptions and caveats of this approach.

To calculate sensitivities at e.g. $1^\circ \times 1^\circ$ resolution from a particular dataset, Eq. (3.8) is applied to all data for a given season and grid-box. An ordinary least squares linear regression fit is then performed through a (conceptual) scatter plot of $\ln \phi$ against $\ln \tau$, where each point represents a single day during the season for which aerosol and cloud data both exist in a particular grid-box. The correlation coefficient (r) and one-sigma error of the regression fit, (ϵ') are also calculated. (This method assumes that a power-law relationship exists between ϕ and τ that can be approximated with $\ln \phi = a + b \ln \tau + \epsilon$, where a and b are constants and ϵ represents errors not accounted for in this linear model, i.e. that $\phi \propto A\tau^b$. See Grandey (2011) for further discussion.)

When moving to larger regions, e.g. $15^\circ \times 15^\circ$, the so-called ‘grid-method’ of Grandey and Stier (2010) is deployed, in contrast to the ‘region-method’ of Quaas et al. (2008, 2009). This means that the value of b (and ϵ') for each $1^\circ \times 1^\circ$ grid-box is calculated first, as described. Then the mean is calculated over the whole $15^\circ \times 15^\circ$ region, with each contributing $1^\circ \times 1^\circ$ - b value being weighted by its $1^\circ \times 1^\circ$ - ϵ' , to give an error-weighted mean over the $15^\circ \times 15^\circ$ region. Seasonal values are then combined with their errors to give an error-weighted annual mean. (Note that the level 3 SSF dataset has a resolution of $2.5^\circ \times 2.5^\circ$, which is then gridded up to $15^\circ \times 15^\circ$ by a similar method.)

Cloud droplet number concentration (N_d)

The left-hand column of Fig. 3.7 shows the annual mean sensitivity, $b = d \ln N_d / d \ln \tau$, for the reference model and each of three satellite datasets. (Annual means are calculated over all seasons, averaged to $15^\circ \times 15^\circ$ weighted by the one-sigma error, using the grid method described above, and then the annual mean is averaged over all four seasons.) White regions indicate where data do not differ statistically from zero at two-sigma confidence. The right-hand column of Fig. 3.7 shows the corresponding correlation coefficient, r . Grey regions denote missing data. For both the model and satellite datasets, regression fits are only carried out between 60° South and 60° North, because satellite retrievals are less reliable over the poles.

The model results show a positive relationship in most regions between N_d and τ , indicating that regions of increasing N_d tend to correspond with regions of increasing aerosol optical depth. Although this does not imply a causal relationship, it does demonstrate that a response consistent with the cloud albedo effect is observed in the model. The strength of this relationship is quantified as an area-weighted mean (AWM) sensitivity of $b = 0.289$ over ocean, and $b = 0.311$ over land, although the correlation coefficient is fairly weak ($r = 0.15$ over ocean and $r = 0.13$ over land). The same positive relationship is seen in all three satellite datasets over ocean, with AWM ranging from $b = 0.107$, $r = 0.05$ for AATSR to $b = 0.179$, $r = 0.17$ for MODIS CERES SSF. So, at least over ocean, the cloud albedo effect in the model is the same sign as in the satellite data, if perhaps too strong. Comparisons between modelled and measured CCN in Sec. 3.5 showed that modelled CCN concentrations are biased low compared to the measurements, especially in marine stratocumulus. Low background values of marine aerosol can mean that anthropogenic aerosol perturbations lead to stronger indirect aerosol effects, because the change in albedo due to any fixed perturbation to N_d increases with decreasing initial N_d . This means that underestimating the background aerosol concentrations can lead to overestimations of $b = d \ln N_d / d \ln \tau$. However, the correlation coefficients from both model and satellites alike are so low, that it would be unwise to place too much emphasis in this analysis on the relative values of the sensitivities themselves.

Over land, the picture from the satellites is more mixed, with AWMs ranging from $b = -0.096$, $r = -0.08$ for AATSR, to $b = 0.125$, $r = 0.11$ for MODIS CERES SSF, which provides no real constraint even to the sign of the relationship. Both AATSR and MODIS Collection 5 mostly show weak negative relationships over land, whereas MODIS CERES SSF shows predominantly positive relationships, like those seen in the model.

Cloud droplet effective radius (r_e)

Following the format of Fig. 3.7, Fig. 3.8 shows the annual mean sensitivity of cloud-top liquid cloud droplet effective radius to aerosol optical depth, $b = d \ln r_e / d \ln \tau$, and the corresponding correlation coefficient, r , for both the reference model and satellite datasets. As expected from Eq. (1.38), the spatial distribution of each of these graphs is approximately the inverse of the corresponding graph in Fig. 3.7, although more regions that are not statistically different from zero are found for r_e than for N_d . Sensitivities are predominantly negative in the model, with a few positive regions, particularly over the tropical Pacific Ocean. Negative sensitivities correspond to the basic model of the cloud albedo effect, and these negative relationships observed in the model indicate that indeed, decreasing droplet size occurs in regions of increasing aerosol in most places. From Eq. (1.38), it is expected that sensitivities for r_e are much smaller than for N_d . This is what is observed: $b = -0.071$

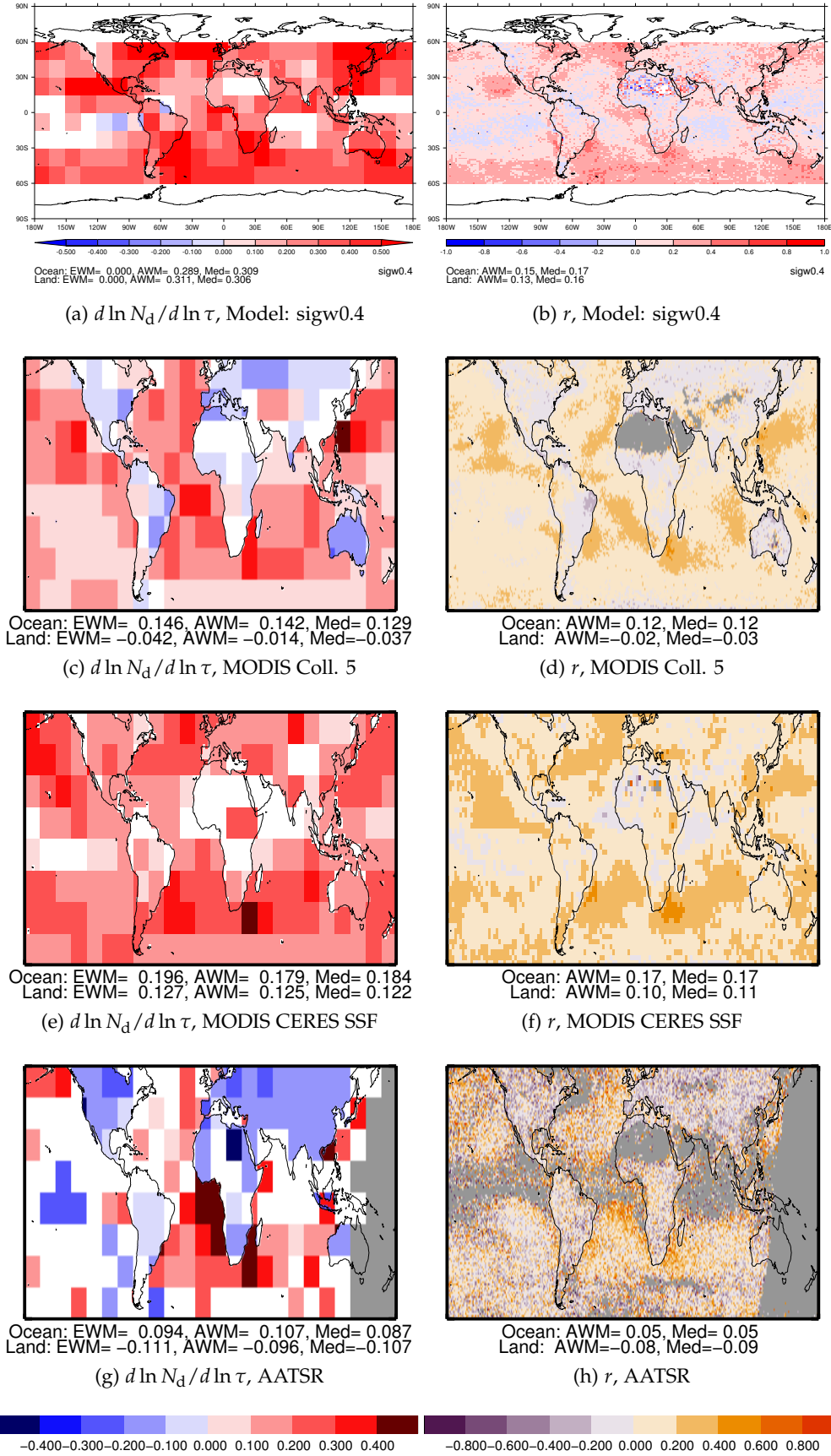


Figure 3.7: Linear regression fits of N_d to aerosol optical depth (τ). Left column shows annual mean (one-sigma error-weighted) gradient of regression slope, $b = d \ln N_d / d \ln \tau$. Right column shows annual mean correlation coefficient r . For: (a,b) reference model sigw0.4; (c,d) Aqua-MODIS Coll. 5; (e,f) Aqua-MODIS CERES SSF ; (g,h) AATSR. White regions show where data do not differ statistically from zero at two-sigma confidence. Grey regions denote missing data. Figures (c–h) of satellite data produced by B. Grandey (Grandey, 2011).

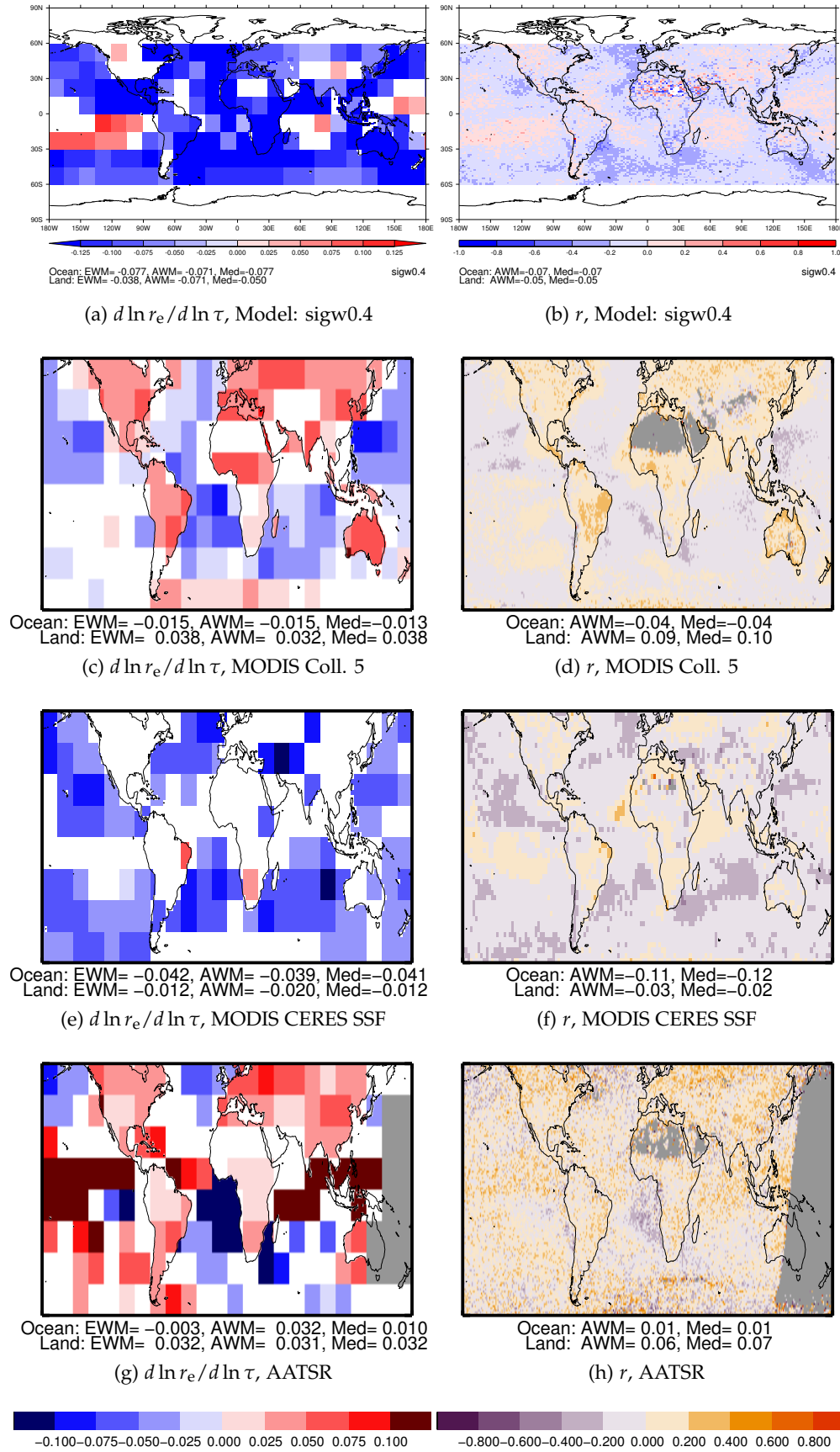


Figure 3.8: For the same model and satellite datasets as Fig. 3.7, linear regression fits of cloud-top liquid cloud droplet effective radius (r_e) to aerosol optical depth (τ). Left column shows annual mean (one-sigma error-weighted) gradient of regression slope, $b = d \ln r_e / d \ln \tau$. Right column shows annual mean correlation coefficient r . Again, figures (c–h) of satellite data produced by B. Grandey (Grandey, 2011).

for r_e over land and ocean, compared to $b \approx 0.3$ for N_d . However, the model correlation coefficients are also very low ($r = -0.07$ over ocean and $r = -0.05$ over land), so there is a lot of scatter about the regression fits.

The satellites provide little constraint over land or ocean, since there are discrepancies over sign in both ocean (AWM ranging from $b = -0.039$, $r = -0.11$ for MODIS CERES SSF, to $b = 0.032$, $r = 0.01$ for AATSR) and land (AWM ranging from $b = -0.020$, $r = -0.03$ for MODIS CERES SSF to $b = 0.031$, $r = 0.06$ for AATSR). These small sensitivities with weak correlation coefficients—and, moreover, conflicting signs—make $d \ln r_e / d \ln \tau$ a rather poor choice of observational constraint on modelling estimates of indirect aerosol effects.

Liquid water path (W)

Again following the format of Fig. 3.7, Fig. 3.9 shows the annual mean sensitivity of liquid water path, W , to aerosol optical depth, $b = d \ln W / d \ln \tau$, and the corresponding correlation coefficient, r , for the reference model and satellite datasets. Here, strong positive relationships are seen in most regions of the model, which correspond to the mechanism of the cloud lifetime effect. Increasing cloud liquid water is indicative of longer cloud lifetime, as the precipitation efficiency is lower when the droplets themselves are more numerous and smaller, in regions of higher aerosol concentrations. Negative regions could indicate an opposite cloud lifetime effect (decreasing liquid water with increasing aerosol) or could indicate that liquid water path is very low compared to the high values of aerosol optical depth, e.g. due to mineral dust in the very dry Saharan dust outflow region. Other than this, negative values are mostly confined to the tropical regions. Since aerosol activation is not properly represented by UKCA-Activate in the convective clouds which dominate the meteorology in the tropics, it is unsurprising that there is little or even negative correlation between the variables in these regions.

Overall, the AWMs from the model are generally strong and positive, with $b = 0.490$, $r = 0.11$ over ocean, and $b = 0.454$, $r = 0.11$ over land. These compare well with the satellite-derived relationships, at least in terms of sign, since all three satellites agree on positive AWMs over both land and ocean, with AWMs ranging over ocean from $b = 0.060$, $r = 0.04$ for MODIS Collection 5, to $b = 0.204$, $r = 0.05$ for AATSR, and over land from $b = 0.137$, $r = 0.10$ for AATSR, to $b = 0.188$, $r = 0.16$ for MODIS CERES SSF. These unanimous positive relationships between all three satellites, provide some validation of the sign of the relationship seen in the model. The magnitude of the effect may be too strong in the model, but with correlation coefficients in the region of 0.1 to 0.2 in all cases, it is difficult to say.

3.8 Conclusions

This chapter has provided a description of the model that will be used for the GCM simulations around which the rest of the work in this thesis is based. The new microphysical aerosol and chemistry model, UKCA, has been summarised briefly, and the implementation of the Abdul-Razzak and Ghan aerosol activation scheme has been described in more detail, since this represents the major technical work underpinning the rest of the thesis. In subsequent chapters, this model will be used to estimate the indirect radiative effects of aerosol. It is therefore important to quantify how well the model simulates CCN, since any error in this value will propagate directly through to errors in cloud droplet number concentration via aerosol activation, and hence to estimates of the indirect aerosol effects. Sec. 3.5 described the evaluation of modelled CCN against measurements in a wide variety

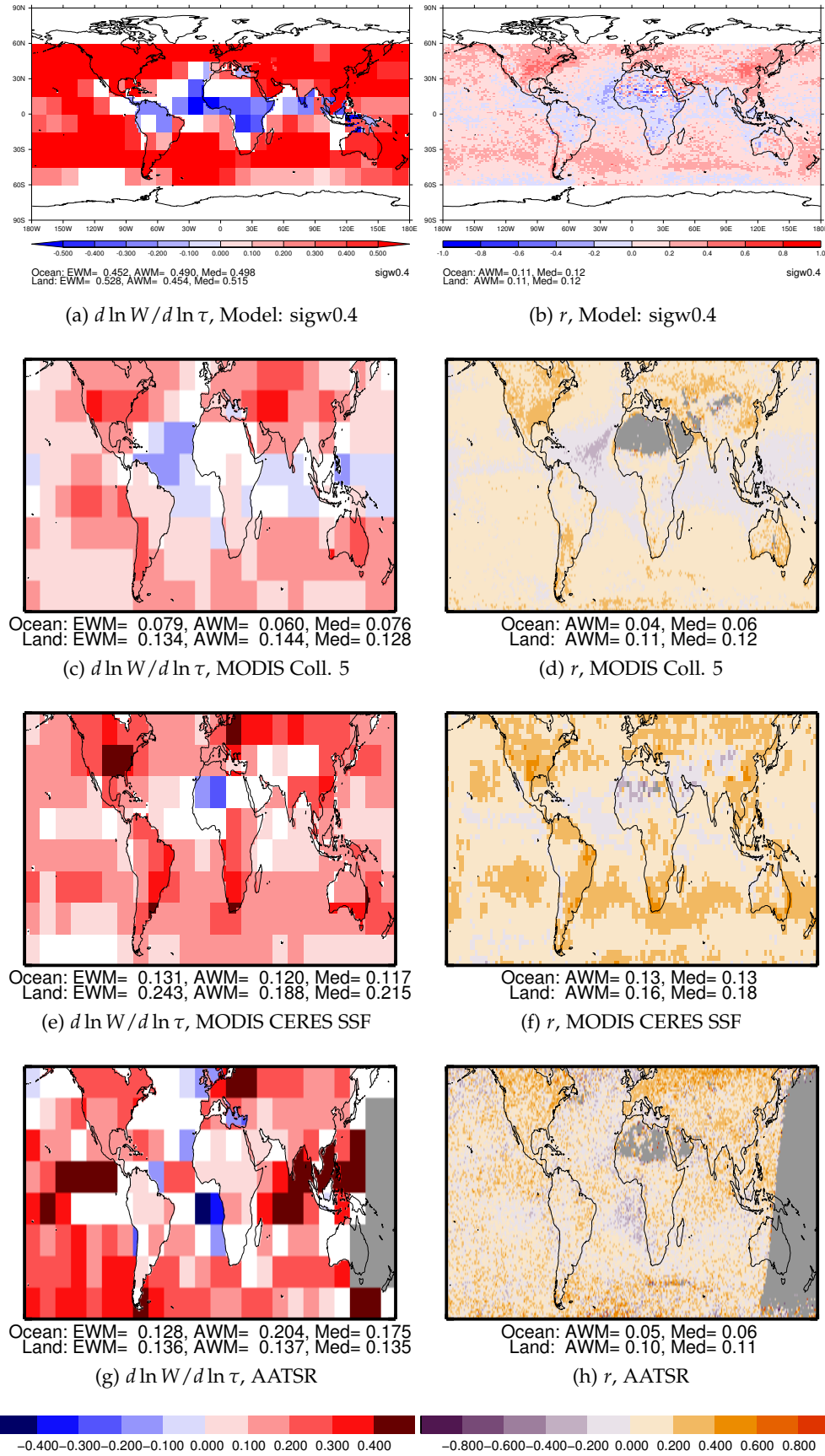


Figure 3.9: For the same model and satellite datasets as Fig. 3.7, linear regression fits of liquid water path (W) to aerosol optical depth (τ). Left column shows annual mean (one-sigma error-weighted) gradient of regression slope, $b = d \ln W / d \ln \tau$. Right column shows annual mean correlation coefficient r . Again, figures (c–h) of satellite data produced by B. Grandey.

of geographical locations, from 55 separate studies in the literature. 90% of modelled CCN were found to fall within a factor of ten of the co-located measured values, with a tendency to be biased low. This is particularly problematic in marine stratocumulus regions, and appears to be especially evident in the stratocumulus of the south-east Pacific.

This lack of CCN in stratocumulus regions is a potential cause of some of the deficiencies in cloud droplet number concentration shown in the evaluation of the model against satellite datasets from MODIS, CERES SSF and AATSR in Sec. 3.6. Annual mean aerosol optical depth (τ), cloud droplet number concentration (N_d), cloud droplet effective radius (r_e), and cloud liquid water path (W), have all been sampled from the reference model at the satellite overpass time, and compared with the same properties retrieved from these satellite instruments. For several of these properties, inter-satellite differences can be larger than satellite–model differences, so the satellites do not provide much constraint on the absolute values obtained by the model. Nevertheless, the three satellites tend to agree on the dominant spatial features of each property, and these have been shown to be well replicated by the model.

Although different datasets may disagree over the absolute values of cloud properties and aerosol optical depth, the concept of the ‘sensitivity’ of one to the other allows aerosol–cloud relationships within a dataset to be quantitatively compared with those found in other datasets. In Sec. 3.7, this method has been used to quantify the sensitivity of aerosol–cloud relationships within a present-day run of the model, which gives some indication of the strength of the indirect aerosol effects within this model. Sensitivities of N_d , r_e and W to τ have been shown for the reference model and three satellite datasets, in Figs. 3.7 to 3.9.

Positive correlations of N_d with τ (consistent with the cloud albedo effect) have been found for the model in most regions, and for all three satellites over ocean, although the slopes are steeper for the model ($b = 0.289$, compared to a range from $b = 0.107$ to $b = 0.120$ from the satellites). Over land, satellites provide conflicting estimates of the sign of the relationship, possibly due to the unreliability of retrievals over land.

As expected, in most regions the model produces negative correlations between r_e and τ , with smaller absolute values than for N_d , due to the inverse power-law relationship between these two cloud properties. However, the satellite datasets are in disagreement over the sign of this relationship particularly over land but also over ocean, and the gradients and correlation coefficients are so small for both model and satellites that this relationship provides a less useful measure than N_d and τ , as a proxy for the cloud albedo effect.

Finally, positive correlations have been found between modelled W , and τ over most regions, with some negative correlations found in dry, dust-dominated and tropical regions with low cloud fractions. Positive correlations between W and τ are consistent with the cloud lifetime effect, and these have also been found over both land and ocean in all three satellite datasets. Gradients of the regression slopes in the model are steeper than those in the satellite datasets, with $b = 0.454$ over land and $b = 0.490$ over ocean (with $r = 0.11$ for both), compared to a range from $b = 0.101$ to $b = 0.204$ from the satellites, with the correlation coefficient ranging from $r = 0.05$ to $r = 0.18$. The satellite datasets have therefore been shown to present a useful constraint on the sign of the sensitivity of W to τ . The relationship in the model was found to have the same sign as the satellites (positive), but with a steeper gradient. This indicates that the cloud lifetime effects may be slightly too strong in the reference model.

In conclusion, comparisons with CCN measurements have shown that HadGEM-UKCA produces CCN concentrations that are within a factor of ten of co-located measured values at the same time and supersaturation. Evaluation of modelled aerosol–cloud relationships against those found in three satellite datasets shows good agreement, at least in the sign

of the relationships. Both model and satellite data show mostly positive relationships between N_d and τ over ocean, and between W and τ over all regions, consistent with the cloud albedo and cloud lifetime effects. If anything, relationships were found to be slightly stronger in the model than the satellite data, but the correlation coefficients of all relationships are small, and this is not necessarily significant.

In terms of improving the representation of aerosol–cloud interactions within the model, sensitivity studies could be performed to assess the relative impacts that some of the assumptions and choices of parameterisations eventually have on modelled estimates of the indirect aerosol effects. For example, one could test the sensitivity to the cloud droplet spectral dispersion relationship, the choice of autoconversion parameterisation, the imposed minimum cloud droplet number concentration, and/or whether it is appropriate on a GCM scale to assume that the number of droplets throughout the vertical is equal to the number of aerosol activated at cloud base. The work in the following chapters focuses on the representation of sub-grid-scale vertical velocity for aerosol activation, and the ultimate effects of the choice of this parameterisation on estimates of the indirect aerosol effects.

Future developments to HadGEM3, such as a two-moment cloud scheme with detailed microphysics and a prognostic cloud droplet number concentration should lead to more physical representation of the indirect aerosol effects within the model. This may hopefully also lead to a reduction in the uncertainty of estimates of the indirect aerosol effects.

Representation of sub-grid-scale vertical velocity

4.1 Introduction

The activation of aerosol to form cloud droplets in the parameterisations described in Chapter 2 is fundamentally dependent on the local supersaturation, which is strongly influenced by the vertical velocity of the rising air [Eq. (1.28)]. Typically, the large-scale vertical velocities resolved at the global circulation model (GCM) grid-scale are small, and it is the unresolved sub-grid-scale fluctuations which give rise to the updraughts associated with cloud formation. It is therefore necessary to account for this sub-grid-scale variability if aerosol activation is to be represented meaningfully in a GCM.

This chapter follows the convention of Golaz et al. (2011) in distinguishing between the local parameterisations of Chapter 2, applicable at cloud-scale, and the sub-grid assumptions necessary to convert these local parameterisations into a grid-box activation parameterisation.

The problem of sub-grid-scale vertical velocity has been addressed in current GCMs by two distinct approaches. One approach uses a probability density function (pdf) to represent the likely distribution of vertical velocity within each grid-box at each timestep, thereby producing a weighted grid-box-average in-cloud droplet number concentration, $\bar{N}_d$, described in Sec. 4.1.1 (Chuang et al., 1997a; Ghan et al., 1997).

An alternative, computationally cheaper approach to capturing the dominant effects of this underlying distribution of vertical velocity is to calculate the number of cloud droplets that would form in the grid-box at a single ‘characteristic’ updraught velocity, w^* . This approach was first used in a GCM by Lohmann et al. (1999), who made the assumption that vertical velocity is dominated by turbulent transport, approximated by the root-mean-square value of the turbulent kinetic energy. Partly due to its simplicity, this method was initially widely adopted by other GCMs (e.g. Takemura et al. (2005), Goto et al. (2008), Ming et al. (2007), Morrison and Gettelman (2008), Gettelman et al. (2008), Wang and Penner (2009)) although most authors concede that it is a considerable simplification compared to the pdf-based approach.

Interest in the concept of characteristic updraught has been recently rekindled in the literature (Morales and Nenes, 2010). However, work later in this chapter will show that

the analytical expressions derived in that study are not necessarily applicable over the full range of aerosol conditions simulated by a GCM.

In this chapter, both of these approaches to the representation of the sub-grid-scale variability of vertical velocity within HadGEM-UKCA are explored, and results are shown from simulations with five different configurations of updraught velocity. In Chapter 5, these results are evaluated by comparing high time-resolution model output with observations from a dozen flight campaigns in a variety of geographic locations.

4.1.1 Mathematical formulation

In the local activation parameterisations described in Chapter 2, the number of activated aerosols is predicted as a function of aerosol properties (size, number and composition), $a_i, i = 1 \dots n$, vertical velocity, w , temperature, T , and pressure, p , such that it can be expressed as $N_a(a_1, \dots, a_n, T, p, w)$. The average number of activated aerosols within a grid-box is denoted as $\overline{N}_a$.

In the **pdf-based approach**, it is assumed that the probability density function of vertical velocity within each grid-box has an explicit shape, $P(w)$, assumed continuous. The grid-box parameterisation is thus determined by calculating the expected value of the local parameterisation at each grid-box:

$$\overline{N}_a = \frac{\int_0^\infty N_a(a_1, \dots, a_n, T, p, w) P(w) dw}{\int_0^\infty P(w) dw}. \quad (4.1)$$

Since aerosol activation does not occur in regions of downdraught, integration is only carried out for $w > 0$.

In the **characteristic approach**, it is assumed that the grid-box parameterisation can be obtained by simply substituting a characteristic vertical velocity, w^* , into the local parameterisation, i.e.

$$\overline{N}_a = N_a(a_1, \dots, a_n, T, p, w^*). \quad (4.2)$$

An obvious motivation for this simplification is the reduction in computational expense by eliminating the integration required by the pdf-based approach at every timestep.

As described in Sec. 3.3, the method by which this average number of activated aerosols, $\overline{N}_a$, (calculated by either method) is then related to the grid-box mean in-cloud droplet number concentration, N_d , is dictated by whether droplet number is treated prognostically or diagnostically by the particular cloud scheme available within the host GCM, as discussed in Sec. 1.5.1.

4.1.2 Probability distributions of vertical velocity

Gaussian distributions

Most models that represent the sub-grid variability of updraught velocity with a pdf assume a Gaussian distribution of probabilities across the grid-box, with mean $\bar{w}$ and standard deviation σ_w , i.e.

$$P(w) = \frac{1}{\sqrt{2\pi}\sigma_w} \exp \left[- \left(\frac{(w - \bar{w})^2}{2\sigma_w^2} \right) \right]. \quad (4.3)$$

Typically, the mean of the distribution, $\bar{w}$, is taken to be the large-scale grid-box mean vertical velocity. This can be either positive or negative, but is usually small, of order

1 cm s^{-1} . (This is equivalent to considering the vertical velocity as a sum of the large-scale velocity, $\bar{w}$, and contributions from the sub-grid-scale variability, w' , such that $w = \bar{w} + w'$.)

In the first studies to employ a Gaussian pdf in a GCM, the distribution was assigned a fixed value of standard deviation, $\sigma_w = 0.5 \text{ m s}^{-1}$, at all grid-points, informed by aircraft observations of stratocumulus cloud off the coast of California (Chuang et al., 1997a). Subsequently, σ_w for stratiform clouds became more commonly related to some measure of turbulence within the model. As discussed by Ghan et al. (1997), many processes can produce sub-grid-scale variability in vertical velocity, but Ghan et al. assume that all sub-grid-scale variability is due to turbulence; σ_w is diagnosed from the turbulent kinetic energy (if predicted by the host GCM) or related to the eddy diffusivity, K , by

$$\sigma_w = \max \left(\frac{\sqrt{2\pi}K}{\Delta z}, 0.1 \right) \text{ m s}^{-1}, \quad (4.4)$$

where Δz is grid-box height. The lower limit of $\sigma_w = 0.1 \text{ m s}^{-1}$ is imposed because turbulence driven by radiative cooling at the cloud-top is poorly resolved above the planetary boundary layer in GCMs with coarse vertical resolution $> 100 \text{ m}$ (Ghan et al., 1997). In subsequent studies, this lower limit was raised to $\sigma_w = 0.2 \text{ m s}^{-1}$ (Ghan et al., 2001b,a; Easter et al., 2004), and to $\sigma_w = 0.3 \text{ m s}^{-1}$ in CAM-Oslo (Storelvmo et al., 2006). In the most recent modelling study that falls into this category, a much higher minimum value of $\sigma_w = 0.7 \text{ m s}^{-1}$ is used in the reference case (Golaz et al., 2011), which is near the upper limit of the measured values of σ_w in the flight campaigns through stratiform cloud considered in Chapter 5.

Deviations from the Gaussian distribution in observations and LES studies

Although some observations show that a Gaussian distribution may be a reasonable approximation for marine stratocumulus cloud (Peng et al., 2005), the sub-grid variability of vertical velocity in other cloud regimes may be better approximated by different forms of $P(w)$. *In situ* observations from intensive aircraft campaigns, as well as longer-term statistics from permanent ground-based remote sensing stations, in different regimes show that variance, skewness and kurtosis of vertical velocity distributions vary not only between different cloud types (Zhu and Zuidema, 2009), but also between clouds of the same type (Moyer and Young, 1991), and even within the vertical profile of a single cloud (Ghate et al., 2010). This makes general parameterisation for use at the grid-scale in GCMs particularly challenging.

Large eddy simulations based on observations of cloud cases representing typical marine and continental boundary layer clouds highlight some of the differences between regimes (Zhu and Zuidema, 2009). Stratocumulus clouds generally have a weakly skewed distribution of updraughts, with varying sign. In shallow cumulus clouds, the dynamic structure of strong narrow updraughts surrounded by broader areas of weaker downdraughts tends to produce strongly positively skewed distributions, and this mechanism is intensified for deep convective systems. These convective clouds are also found to have extremely large kurtosis of variables within the cloud layer. The difficulty in replicating this shape of pdf may be a bottleneck for the future development of pdf-based cloud parameterisations.

The sign ambiguity noted in the stratocumulus simulations is also borne out in measurements: both positive and negative skewness have been observed in marine stratocumulus in the north-east Atlantic (Guibert et al., 2003), and in the stratocumulus deck off the coast of California (Moyer and Young, 1991). Positive skewness means that the probability distribu-

tion has a longer tail for updraughts than for downdraughts, indicating that there are fewer updraughts, but that these are stronger than the strongest downdraughts. The latter study presents observations from three flights through the same cloud deck, that each display very different vertical profiles of $P(w)$, skewness and kurtosis. Convective turbulence within a marine-stratocumulus topped boundary layer is potentially influenced not only by the surface buoyancy flux, but also by radiative cooling at the cloud-top (Nicholls, 1984)) (This physical process can be replicated in large eddy simulations of marine stratocumulus cases (Zhu and Zuidema, 2009).) The turbulent structure of the stratocumulus-topped boundary layer is also affected by cloud-top entrainment, phase of diurnal cycle, latent heat exchange, and whether or not the stratocumulus layer overlies cumulus clouds, which may or may not extend up into the stratocumulus deck. To complicate matters further, all of these contributing factors can be interdependent.

Large eddy simulations have helped to improve understanding of the physical processes that cause skewness of vertical velocity distributions, but further observations are still required. LES studies of the south east Pacific marine stratocumulus by Mechem et al. (2012), showed that positively skewed distributions of vertical velocity typically occur when the circulation is dominated by surface buoyancy driven convection. In earlier LES studies, Golaz et al. (2005) concluded that negatively skewed distributions are caused by negatively buoyant downdraughts, driven by both radiative cooling at the cloud-top and evaporative cooling (Nicholls, 1989; Yamaguchi and Randall, 2008).

Continental stratocumulus are more spatially and temporally variable than marine stratocumulus which makes them even harder to parameterise. However, some studies have taken advantage of long term statistics, obtained from ground-based Doppler cloud radar instruments at the Atmospheric Radiation Measurement Program's Southern Great Plains observing facility near Lamont, Oklahoma (Ghate et al., 2010; Tonttila et al., 2011). In general, these show $P(w)$ to be weakly positively skewed in the cloud layer, and negatively skewed near the cloud-top but, as for the marine case, the signs can vary from cloud to cloud. This is shown to be linked to the intensity of the surface buoyancy flux, but a definitive explanation of the underlying mechanisms would require these measurements to be considered in conjunction with large eddy simulation (LES) studies (ongoing work, see e.g. Ghate et al. (2010)).

Outlook for the pdf-approach

In summary, the exact functional form of $P(w)$ varies depending on cloud regime, boundary layer type and diurnal cycle (amongst other factors), and the processes governing this shape are far from understood and hence difficult to parameterise. Nevertheless, ongoing developments are underway to improve on the basic, initial approximation of a Gaussian pdf. For instance, Larson et al. (2002) used a combination of aircraft data and LES to show that a double (binomial) Gaussian pdf provides the best representation of sub-grid-scale variability in boundary layer cloud, out of five families of analytical pdfs tested, when compared to observed distributions from stratocumulus, cumulus and clear boundary layers measured during two flight campaigns (Albrecht et al., 1988, 1995). Following on from this, a turbulence cloud parameterisation based on the double Gaussian pdf has been developed (Golaz et al., 2002, 2007) and has recently been extended to the prediction of cloud droplet number in the single-column version of the GFDL GCM (Guo et al., 2010) but, at the time of writing, this approach is yet to be implemented in a GCM.

An alternative approach may be to use a nested modelling framework, in which a LES or cloud-resolving model could be directly activated by GCMs in the most important

boundary layer cloud-forming regions (Zhu et al., 2010). Currently, this approach is in the early development stages, but with potential increases in computational power in the future, it may prove to be useful.

4.1.3 Characteristic updraught velocity

The choice of characteristic updraught velocity itself, w^* , is obviously of critical importance for models which utilise it. The first use of characteristic updraught velocity in a model with prognostic droplet number was made by Lohmann et al. (1999), in which w^* was defined as

$$w^* = \bar{w} + c\sqrt{\text{TKE}}, \quad (4.5)$$

where $\bar{w}$ is the large-scale grid-box mean vertical velocity, TKE is the turbulent kinetic energy and $c = 0.7$ is an empirically derived factor. (This formulation is followed by Takemura et al. (2005) and Goto et al. (2008).) Lohmann (2002) increased the parameter c for stratiform clouds to $c = 1.33$ and extended the parameterisation to also apply to convective clouds, with $c = 0.5$ and TKE replaced by convective available potential energy (CAPE) in Eq. (4.5) (although this extension for convective clouds has since been retracted in later versions of ECHAM). Ming et al. (2007) follow a similar method for convective clouds.

An alternative approach, taken by Morrison and Gettelman (2008) and Gettelman et al. (2008), is to derive w^* directly from the eddy diffusivity and a constant characteristic mixing length, $l_c = 30$ m, i.e.

$$w^* = \max\left(\frac{K}{l_c}, 0.1\right) \text{ m s}^{-1}. \quad (4.6)$$

Wang and Penner (2009) use an amalgamation of both the Lohmann et al. (1999) and Morrison and Gettelman (2008) formulations outlined above:

$$w^* = \bar{w} + c(\sigma_w) \times \sigma_w, \quad \text{where } \sigma_w = \max\left(\frac{K}{l_c}, 0.1\right) \text{ m s}^{-1}, \quad (4.7)$$

where the mixing length varies from $l_c = 300$ m in the boundary layer to $l_c = 30$ m in the free atmosphere. The functional form of $c(\sigma_w)$ is determined from an empirical fit, such that the predicted cloud droplet number approximately matches the number obtained by integration over a normal distribution of zero mean and standard deviation σ_w (a concession to the pdf-based approach outlined above). This fitting procedure leads to values of $c(\sigma_w)$ between 0.2 and 0.6. (Wang and Penner (2009) report an accuracy of 10 %, provided that the accumulation mode aerosol concentration is less than 1000 cm^{-3} .)

Most recently, the concept of characteristic updraught has been re-addressed by Morales and Nenes (2010). In this context, the characteristic updraught velocity, w^* , is defined to be the value of w that yields a cloud droplet number concentration representative of what would be obtained by taking the pdf-based approach (i.e. w^* is the value of w for which $N_a(w^*) = \bar{N}_a$). This paper proposes the use of analytical formulations of w^* based on the Twomey (1959) droplet parameterisation for the computation of average values of CDNC over a Gaussian pdf of updraught. These are shown to agree well with numerical integrations using the Fountoukis and Nenes (2005) aerosol activation parameterisation for a limited number of test cases.

However, in Sec. 4.2.2, we explore this approach in a wider range of test cases with both box-model and GCM simulations, and find that the simple expressions for w^* proposed

here are formulated in terms of a parameter which is sensitive to the aerosol properties (size, number and composition) needed to calculate it. The expressions for w^* are therefore not suitable to be applied across the wide range of aerosol variability encountered with a full global climate simulation.

4.2 Baseline configuration of UKCA-Activate

As described in Sec. 3.3, HadGEM3-UKCA uses a single-moment cloud scheme (Wilson et al., 2008), in which cloud droplet number concentration is a purely diagnostic quantity. At each time-step, the in-cloud droplet number concentration for each grid box (N_d) is simply a direct function of the expected number of aerosols that could activate ($\overline{N_a}$), diagnosed from the grid-box aerosol distribution and meteorological conditions at that instant. Aerosols that are activated in a grid-box at one time-step are available for activation again at the next time-step, unless they have been transported away or removed by other sink processes.

The baseline configuration of UKCA-Activate uses the pdf-based representation of the sub-grid variability of vertical velocity (results of box-model tests outlined below indicate reasons for not choosing the characteristic updraught approach). Despite the previous discussion relating to the likely skewed pdf of updraught in certain cloud types (Sec. 4.1.2), this work uses a Gaussian distribution, as in Eq. (4.3). The studies which follow explore the effects of a range of fixed values of σ_w , as well as an experiment in which σ_w is derived from the model turbulent kinetic energy. Currently, measurements of vertical velocity statistics are so geographically and temporally sparse that even if a more complex functional form of $P(w)$ were deployed, its usefulness could only be informed by a very limited set of measurements. It would be difficult to evaluate whether it constituted a global improvement compared to observations, other than in the very limited temporal and spatial regime of such observations. For this reason, the added complexity of higher moments of $P(w)$ is not yet justifiable in a GCM, and this work will focus on the first and second moments alone. A more extensive, co-ordinated measurement effort, in conjunction with further LES studies, would be required to make further progress on the parameterisation of sub-grid vertical velocity variability for GCMs.

4.2.1 Box-model tests for number of bins, extent of distribution

Within a GCM, the calculation of $\overline{N_a}$ using the pdf-based approach requires numerical integration in every grid-box at every timestep. Eq. (4.1) is discretised as

$$\overline{N_a} = \frac{\sum_{i=0}^n N_a(w_i) P(w_i) \Delta w_i}{\sum_{i=0}^n P(w_i) \Delta w_i}, \quad (4.8)$$

where n is the total number of updraught bins and Δw_i is the width of each bin. In Eq. (4.1), integration is defined over the range $0 < w < \infty$, but in practice a finite upper limit is required. For a Gaussian pdf, it is reasonable to consider this upper limit in terms of the standard deviation i.e. $w_{\max} = \alpha \sigma_w = \sum_{i=1}^n \Delta w_i$. Given the limit on computing resources and time, there is a balance to be struck between the number of bins requested, n , and the range of the pdf that they cover and over which they are shared. For linearly-spaced bins, doubling the range while keeping the number of bins constant obviously doubles the bin width. This provides coverage of a greater proportion of the pdf at a cost of reduced resolution of its shape. As illustrated in the box-model example shown in Fig. 4.1, this would have most effect at low w where $N_a(w)$ (green asterisks) has the steepest gradient. The quantity being integrated is actually $N_a(w) \times P(w)$ (denoted by red diamonds in

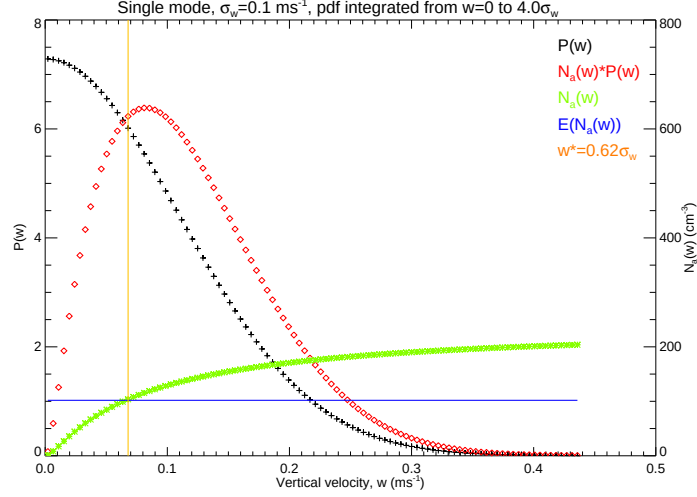


Figure 4.1: Results from running the ARG box-model with a single mode of ammonium sulphate aerosol. On LH vertical axis, black ‘plus’ signs show $P(w_i)$ with $\sigma_w = 0.1 \text{ m s}^{-1}$ for 100 bins of w , ranging from $0 < w < 4\sigma_w$. On RH vertical axis, green asterisks show $N_a(w_i)$; red diamonds show $N_a(w_i) \times P(w_i)$ and blue line shows $E(N_a(w)) = \bar{N}_a$.

Fig. 4.1), and it is tempting to suppose that the bin width ought to be proportional to some derivative of it. However, since the functional form of $N_a(w)$ over a range of w is non-analytic, the derivative of $N_a(w) \times P(w)$ cannot be taken before each value of $N_a(w_i)$ is calculated and that calculation would require Δw_i to be defined *a priori*. Clearly the derivative cannot be used to calculate the bin width on which it is dependent. This could be solved by calculating $N_a(w)$ twice; first a small number of linearly spaced bins could be used to determine the approximate shape of $N_a(w)$, then the appropriate derivative could be taken numerically and hence used to define the bin-widths for the second time that $N_a(w)$ is calculated, this time with a greater number of bins. However, due to the smooth, monotonically-increasing shape of $N_a(w)$ in this case, the gain would be marginal for the cost, so the bins are simply spaced linearly.

Fig. 4.1 illustrates the expected value calculation with the ARG box-model (described in Sec. 2.2), run with a single log-normal mode of totally soluble ammonium sulphate ($r = 0.5 \mu\text{m}$, $\sigma_g = 2$, $N_{\text{conc}} = 230 \text{ cm}^{-3}$). The value of $P(w)$ with $\sigma_w = 0.1 \text{ m s}^{-1}$, calculated at each of 100 equally-spaced values of w , extending from $w = 0$ to $4\sigma_w$, is shown with black plus signs, corresponding to the scale on the left hand vertical axis. The value of N_a , calculated at each value of w , is shown in green asterisks, and corresponds to the scale on the right hand vertical axis. For illustration purposes, the product $N_a(w) \times P(w)$ is shown with red diamonds. The expected value of N_a is calculated using Eq. (4.8), and shown as a blue horizontal line that relates to the right hand vertical axis. The vertical yellow line indicates the value of w for which $N_a(w^*) = \bar{N}_a$, which defines the characteristic value of updraught, w^* .

Box-model convergence tests were carried out in order to optimise α and n . $\bar{N}_a$ was calculated as previously described with the AR&G box-model, over the ranges $0 < w < \alpha\sigma_w$ for $\alpha \in 1, 2, 3, 4, 5, 10$, with number of bins (n) taking ten values between 5 and 1000, at ten logarithmically-spaced values of σ_w ranging from 0.1 to 2.0 m s^{-1} , for each of four multi-modal aerosol test cases, representing typical background, urban, continental and

Test case	Mean radius [μm]		Number [cm^{-3}]		Sol. mass frac.		λ		
	Mode 1	Mode 2	Mode 1	Mode 2	Mode 1	Mode 2	$\lambda_{0.1}$	$\lambda_{1.9}$	$\bar{\lambda}$ (lin. fit)
1	5.0	5.0	100	100	1	0.1	0.62	0.54	0.53
2	0.05	5.0	100	100	1	1	0.50	0.22	0.19
3	0.05	0.5	100	100	1	1	0.50	0.62	0.65
4	0.05	0.5	100	100	1	0.1	0.58	0.50	0.49
5	0.05	5.0	100	100	1	0.1	0.50	0.26	0.24
6	0.05	0.5	1000	100	1	1	0.58	0.70	0.70
7	0.05	0.5	1000	100	0.33	1	0.54	0.70	0.73

Table 4.1: Aerosol mode configurations for box-model tests. $\lambda_{0.1}$ refers to λ calculated with $\sigma_w = 0.1 \text{ m s}^{-1}$, and $\lambda_{1.9}$ with the tenth value of $\sigma_w = 1.9 \text{ m s}^{-1}$.

marine conditions (from Whitby (1978)). The percentage difference in $\bar{N}_a$ from benchmark examples for each case with 100,000 linearly-spaced bins over the range $0 < w < 10\sigma_w$ was calculated for each of these combinations of α and n . Optimal values were found to be $n = 20$ bins over the range $0 < w < 4\sigma_w$, which gives results to within 2 % of those obtained in the benchmark example, in all four aerosol test cases.

4.2.2 Box-model exploration of characteristic updraught

In this configuration, the ARG box-model has been used to explore the form of characteristic updraught proposed by Morales and Nenes (2010). This paper defines the characteristic updraught velocity, w^* , to be the value of w for which $N_a(w^*) = \bar{N}_a$. Essentially, an analytical formulation of w^* is proposed, based on the Twomey (1959) approximation which assumes a power law dependence of the CCN spectrum, i.e. $N_a = cS^k$. The characteristic updraught is expressed as $w^* = \lambda\sigma_w$, and an analytical expression is derived to give λ in terms of k (the steepness of the CCN spectrum).

We have conducted tests with the box-model to explore the relationship between w^* and σ_w , and to investigate the range of values that λ might take, and hence whether this approach is useful for implementation in a GCM.

In each test case, the box-model was run with two modes of aerosol, both with a fixed geometric standard deviation of 2. The geometric mean radius, aerosol number concentration and soluble mass fraction (ratio of soluble ammonium sulphate to insoluble core with same MMR) of each mode, for each test case, are listed in Table 4.1. As described above, the box-model was run 100 times to calculate $N_a(w)$ at 100 equally-spaced values of w over the range $0 < w < 4\sigma_w$. For each aerosol test case, this was repeated with ten equally-spaced values of $\sigma_w = 0.1, 0.3, \dots, 1.9 \text{ m s}^{-1}$. For each, the expected value was then calculated for a Gaussian pdf of updraught velocity with mean $\bar{w} = 0$ and standard deviation σ_w , to yield $\bar{N}_a(\sigma_w)$. Next, w^* was identified by finding the closest bin of w which yields $N_a(w^*) = \bar{N}_a$, and λ calculated from $w^* = \lambda\sigma_w$. For each test case, $\lambda_{0.1} = \lambda(\sigma_w = 0.1 \text{ m s}^{-1})$ and $\lambda_{1.9} = \lambda(\sigma_w = 1.9 \text{ m s}^{-1})$ are shown in Table 4.1. w^* was plotted for each of the ten values of σ_w , and a linear fit was performed, the gradient of which gives an average value of λ over the range of σ_w for each test case. This is listed in the final column of Table 4.1.

The results of these tests show that λ is highly aerosol-dependent and is affected by the composition, number and size of aerosol particles. The value of λ for a given aerosol test case also varies widely depending on the value of σ_w . While Morales and Nenes state that the value of λ can vary from $\lambda = 0.55$ to 0.7 (for $k = 0.1$ (clean) to $k = 1.5$ (polluted))

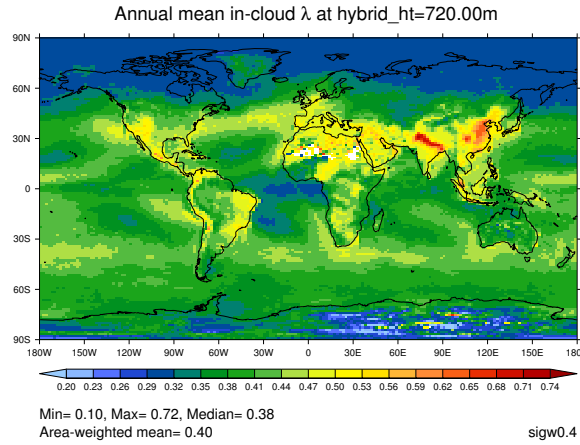


Figure 4.2: Annual mean in-cloud λ calculated from $w^* = \lambda \sigma_w$ from model configuration sigw0.4 at hybrid height= 720 m.

conditions), these tests have shown λ to be as low as 0.19 in the box model studies. The next section shows the typical values of w^* and λ that occur in a GCM.

GCM examples of w^* and λ

The values of λ relevant to a GCM can be calculated by using a pdf of updraught to calculate $\overline{N_d}$ at each grid-point within the GCM, but also diagnosing what the characteristic updraught would need to have been at each grid-point to give the expected cloud droplet number concentration obtained with the pdf. λ can then be calculated from the known (either prescribed or diagnosed) value of σ_w . Fig. 4.2 shows the annual mean of λ from the reference case of HadGEM-UKCA, run with a pdf of updraught with a fixed width of $\sigma_w = 0.4 \text{ m s}^{-1}$. Even in the annual mean, λ is highly spatially variable, and takes on values from $\lambda = 0.1$ to 0.72, with an area-weighted mean value of 0.4. We note that this is considerably lower than the values that Morales and Nenes suggest might be appropriate ($\lambda = 0.65$ derived using the Twomey expression for N_a or $\lambda = 0.68$ derived from Fountoukis and Nenes (2005)). Given the variable spatial (and temporal, not shown) nature of λ derived from the GCM with a full pdf, it seems inappropriate to assume that a fixed value of λ would be suitable for use in a GCM. Further, tests indicate that only 20 bins of updraught are required to faithfully describe the full pdf, and so abandoning the λ -based approach would not be computationally extortionate. Use of a characteristic updraught would seem to underestimate the potential effect of the non-linear relationship between w^* and CDNC, and hence add unnecessary extra uncertainty to estimates of the indirect aerosol effects.

4.2.3 Pdf-based approach: estimation of σ_w from turbulent kinetic energy

Following the method of Ghan et al. (1997) described in Sec. 4.1.2, we make the assumption that all sub-grid-scale variability in vertical velocity is due to turbulence. HadGEM3 uses the Lock et al. (2000) boundary layer turbulent mixing scheme. This scheme combines non-locally determined eddy-diffusivity profiles for turbulence driven by both surface fluxes and cloud-top processes (radiative and evaporative cooling), with an explicit parameterisation for the entrainment rate (derived by Lock (1998)). Within the scheme, different mixing regimes associated with seven different boundary layer types need to be identified so that mixing parameterisations can be selected accordingly. Unstable layers are identified first,

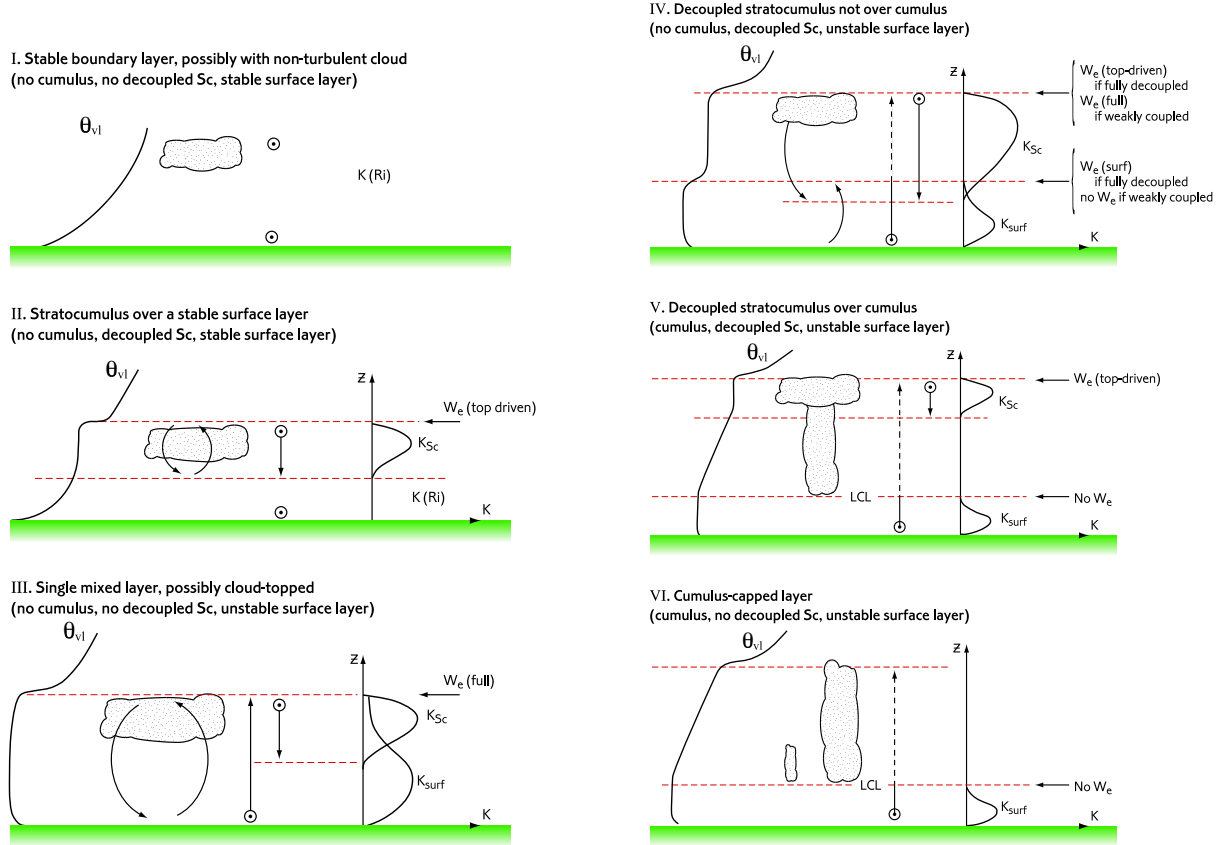


Figure 4.3: Schematic representation of boundary layer types 1 to 6 (reproduced with permission from Lock and Edwards (2011), © Crown copyright 2011, the Met Office).

by consideration of the buoyancy of undiluted parcels lifted from the surface or lowered from the top of a layer cloud. Well-mixed layers (clear and stratocumulus-topped) can then be distinguished from those in which cumulus convection is present and appropriate mixing schemes can then be used for each. The well-mixed layers use a development of an established non-local eddy-viscosity-based mixing scheme (Holtslag and Boville, 1993), modified to include the effects of turbulence driven by radiative and evaporative cooling at the cloud top as well as surface-driven turbulence. Cumulus layers are parameterised using a mass-flux convection scheme (Gregory and Rowntree, 1990). Full details are given by Lock et al. (2000). Finally, parcel descent is used to calculate the depth of turbulent mixing driven from cloud-top. This allows the diagnosis of decoupled stratocumulus layers or even for cloud-top driven turbulence to extend right down to the surface.

In summary, the scheme identifies seven different types of boundary layer that are distinguished by the relative positions of stable and unstable turbulently-mixed layers and cumulus cloud layers, six of which are shown in 4.3. (Type 7 indicates a shear-dominated layer, not depicted.)

Turbulent kinetic energy (TKE) is diagnosed from the modelled eddy diffusivity profiles as

$$\text{TKE} = \left(\frac{K}{l} \right)^2, \quad (4.9)$$

Run name	Vertical velocity in levels 1 to 11	Levels 12 to 38
sigw0.1	$P(w), \sigma_w = 0.1 \text{ m s}^{-1}$	$\sigma_w = 0.1 \text{ m s}^{-1}$
sigw0.4	$P(w), \sigma_w = 0.4 \text{ m s}^{-1}$	$\sigma_w = 0.4 \text{ m s}^{-1}$
sigw0.7	$P(w), \sigma_w = 0.7 \text{ m s}^{-1}$	$\sigma_w = 0.7 \text{ m s}^{-1}$
TKE_0.1	$P(w), \sigma_w = \max\left(\sqrt{\frac{2}{3}\text{TKE}}, 0.1\right) \text{ m s}^{-1}$	$\sigma_w = 0.1 \text{ m s}^{-1}$
w_0.32	No pdf, $w = 0.32 \text{ m s}^{-1}$ everywhere	$w = 0.32 \text{ m s}^{-1}$

Table 4.2: Configurations of vertical velocity used in HadGEM-UKCA simulations.

where K is the eddy diffusivity coefficient and l the mixing length. In the HadGEM3 boundary layer scheme, l is a combination of boundary layer depth and height above the surface, dependent on the type of boundary layer (full technical details are given by Lock and Edwards (2011); Lock et al. (2000)). The boundary layer scheme operates in the lowest ~ 2.5 km of the model troposphere (levels one to eleven inclusive), so in some cases, turbulence can be generated above what is strictly defined as the planetary boundary layer. In the free atmosphere, the model contains no physical way of calculating a mixing length, so a fixed value of 40 m is used.

Turbulent kinetic energy per unit mass is defined as $\text{TKE} = \frac{1}{2} (\sigma_u^2 + \sigma_v^2 + \sigma_w^2)$. In the base-line configuration of UKCA-Activate, isotropic turbulence is assumed such that $\sigma_u = \sigma_v = \sigma_w$, and hence $\text{TKE} = \frac{3}{2} \sigma_w^2$. Therefore, within the scope of the boundary layer scheme, σ_w is diagnosed as

$$\sigma_w = \max\left(\sqrt{\frac{2}{3}\text{TKE}}, \sigma_w^{\min}\right) \text{ m s}^{-1}. \quad (4.10)$$

In levels 12 to 38, turbulent kinetic energy is not diagnosed by the model, and σ_w must take on a fixed value, chosen here to be $\sigma_w^{\min} = 0.1 \text{ m s}^{-1}$.

4.3 The impact of different vertical velocity configurations on cloud properties

To assess the effects of a) the choice of σ_w in a Gaussian pdf and b) the pdf-based approach compared to a single fixed value of vertical velocity, UKCA-Activate has been run with five different configurations of vertical velocity, outlined in Table 4.2. In the first three configurations, **sigw0.1**, **sigw0.4** and **sigw0.7**, the value of σ_w is fixed at 0.1, 0.4 and 0.7 m s^{-1} . This allows us to consider the effect of different widths of $P(w)$ on $\overline{N_d}$, in isolation from the effects of the variation of turbulence in the boundary layer scheme. These values broadly cover the range of the majority of measured σ_w from the flight campaign datasets that will be used for the model evaluation in Chapter 5, although values of σ_w greater than 0.7 m s^{-1} were recorded in several stratiform cases (shown later, in Fig. 5.15). Values of σ_w less than 0.1 m s^{-1} were only recorded on two individual flights. The median value of all of the measurements in the database is close to 0.4 m s^{-1} , and so model run sigw0.4 is used as the reference case in the comparisons between different model configurations which follow.

Model run **TKE_0.1** follows the configuration described in Sec. 4.2.3, i.e. $P(w)$, with $\sigma_w = \max\left(\sqrt{\frac{2}{3}\text{TKE}}, 0.1\right) \text{ m s}^{-1}$.

Finally, the pdf is abandoned for the characteristic updraught approach, and the in-cloud vertical velocity is set to a single fixed value everywhere. The characteristic value chosen is the mean updraught, w_+ , of the reference case sigw0.4, where

$$w_+ = \frac{\int_0^\infty wP(w)dw}{\int_0^\infty P(w)dw}. \quad (4.11)$$

As shown in Fountoukis et al. (2007), for a Gaussian pdf [Eq. (4.3)] with zero mean velocity, this cancels down to

$$w_+ = \frac{\frac{1}{\sqrt{2\pi}\sigma_w} \int_0^\infty w \exp\left[-\left(\frac{(w-\bar{w})^2}{2\sigma_w^2}\right)\right] dw}{1/2} = \sqrt{\frac{2}{\pi}}\sigma_w \approx 0.8\sigma_w. \quad (4.12)$$

So for $\sigma_w = 0.4 \text{ m s}^{-1}$, Eq. (4.12) gives $w_+ = 0.32 \text{ m s}^{-1}$ and hence the model configuration is referred to as **w_0.32**. In this configuration, the aerosol activation scheme only has to run once (with $w = w_+$) at each grid-point at each time-step.

4.3.1 Model configuration intercomparison

Fig. 4.4 to 4.14 illustrate how differences between these different model configurations of vertical velocity manifest themselves in the annual mean fields of CDNC, effective radius (r_e), and liquid water path (LWP). Fig. 4.4 shows annual mean global fields of each of these cloud properties, for model configuration sigw0.4, which will be used as a reference case.

Each subsequent set of figures (4.5 to 4.7) comprises annual means of the cloud properties considered in turn, for the other model configurations listed in Table 4.2, alongside the difference between each of these configurations and sigw0.4.

To summarise, Fig. 4.8 shows bar charts of the difference between the annual, global, area-weighted mean of each of these properties from each model configuration with respect to sigw0.4. For TKE_0.1, differences are also shown with respect to sigw0.1.

Note that annual means of cloud properties do not include contributions from grid-points that contained no liquid cloud at a given timestep. These points are treated as missing values in the time averages, and ignored. A grid-point is defined as cloudy if both the liquid cloud fraction and the cloud liquid water content are greater than zero, i.e. if $f_c > 0$ and $q_{cl} > 0$. Some properties are referred to as ‘in-cloud’, to emphasise that, say, the cloud droplet number concentration is the average concentration of droplets in the cloudy fraction of that grid-box, not the average droplet concentration if the cloud were spread out over the whole volume of the box. Note, however, that contributions to the annual means from cloudy grid-points are not weighted by cloud fraction, so a grid-point with 1% cloud cover has equal weighting in the mean to a point with 100% cloud cover.

Cloud droplet number concentration

Fig. 4.5 shows annual mean in-cloud droplet number concentration at cloud-top for each of model configurations sigw0.1, sigw0.7, TKE_0.1 and w_0.32, and the difference of each of these with respect to sigw0.4. It is clear from the sigw0.1 and sigw0.7 rows, that changing the variance of $P(w)$ can have a significant impact on CDNC at cloud top. The sigw0.1 run shows very low values of CDNC, compared to sigw0.4. The magnitude of the difference is greatest in regions of high CCN (e.g. over China) where increasing the width of the vertical velocity pdf allows considerably more aerosols to activate. (Increasing σ_w increases the weighting toward higher values of vertical velocity in each grid-box, which increases

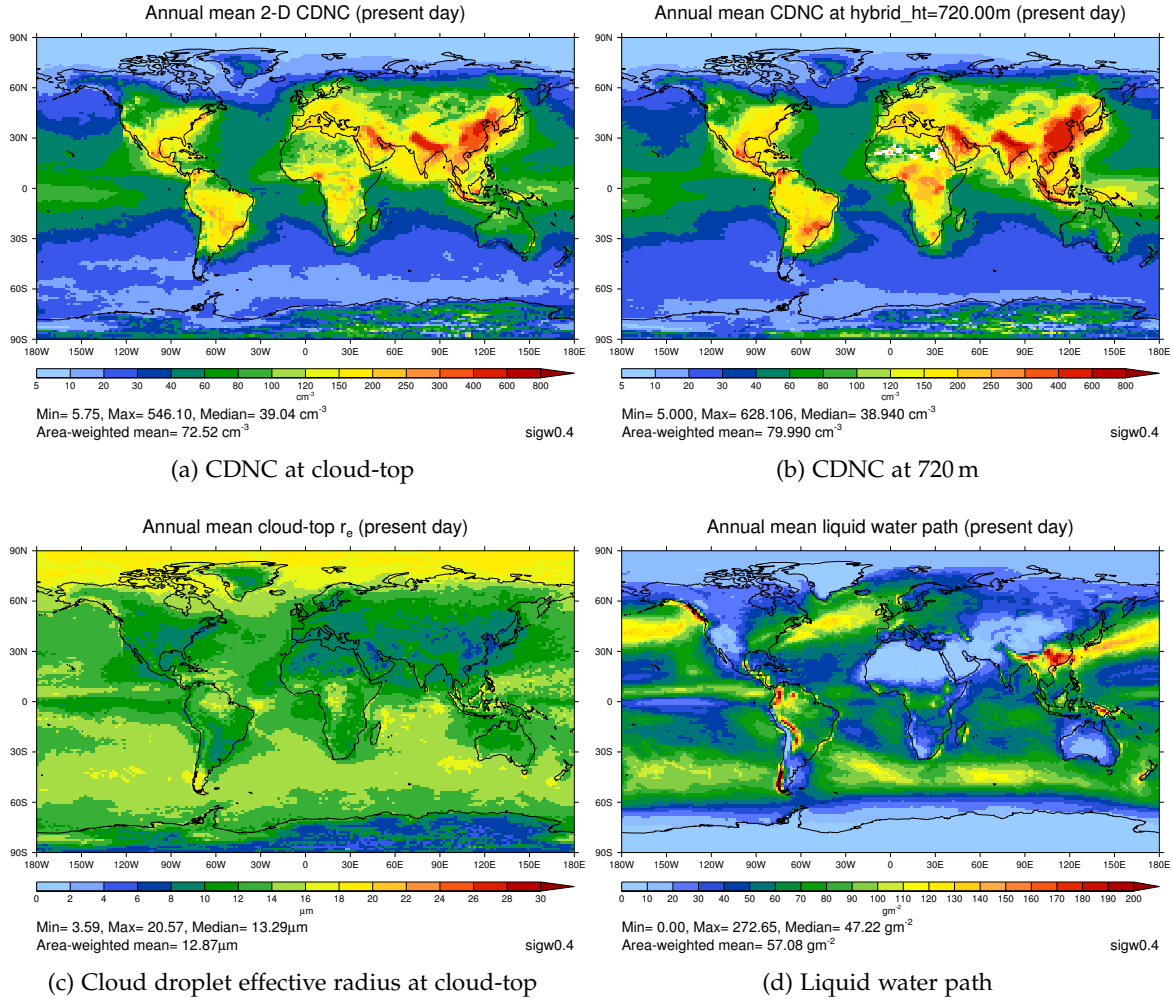


Figure 4.4: Annual mean (a) in-cloud droplet number concentration at cloud-top, (b) same (CDNC) at hybrid height = 720 m, (c) liquid cloud droplet effective radius at cloud-top, (d) liquid water path. All from model configuration sigw0.4. Note non-linear colour scales on (a) and (b).

$S_{\max}$ and hence reduces $a_{\min}$. Since the number distribution of aerosols in each mode is log-normal, decreasing the size of the smallest particle that can activate has an exponential effect on the total number activated).

Furthermore, the difference between the sigw0.1 and sigw0.7 rows—and in particular the difference in magnitude of the difference plots of each of these cases from sigw0.4—illustrates the non-linearity of this effect. A decrease of $\sigma_w = 0.4$ to 0.1 m s^{-1} leads to a global area-weighted mean decrease of CDNC (a change of -27.5 cm^{-3}) compared to a global mean increase (a change of $+9.72 \text{ cm}^{-3}$), only a third as large, when σ_w is increased by the same amount from $\sigma_w = 0.4$ to 0.7 m s^{-1} . Over land, the effect is far more pronounced. This non-linearity in the response is due to a levelling-out of the fraction of activation with increasing supersaturation. Once the fraction of activation is close to 1, further increases in σ_w cease to have an effect.

The difference plot in the third row of Fig. 4.5 shows the effect that using TKE-derived σ_w has on CDNC, compared to sigw0.4. Overall there is a global mean decrease in cloud-top

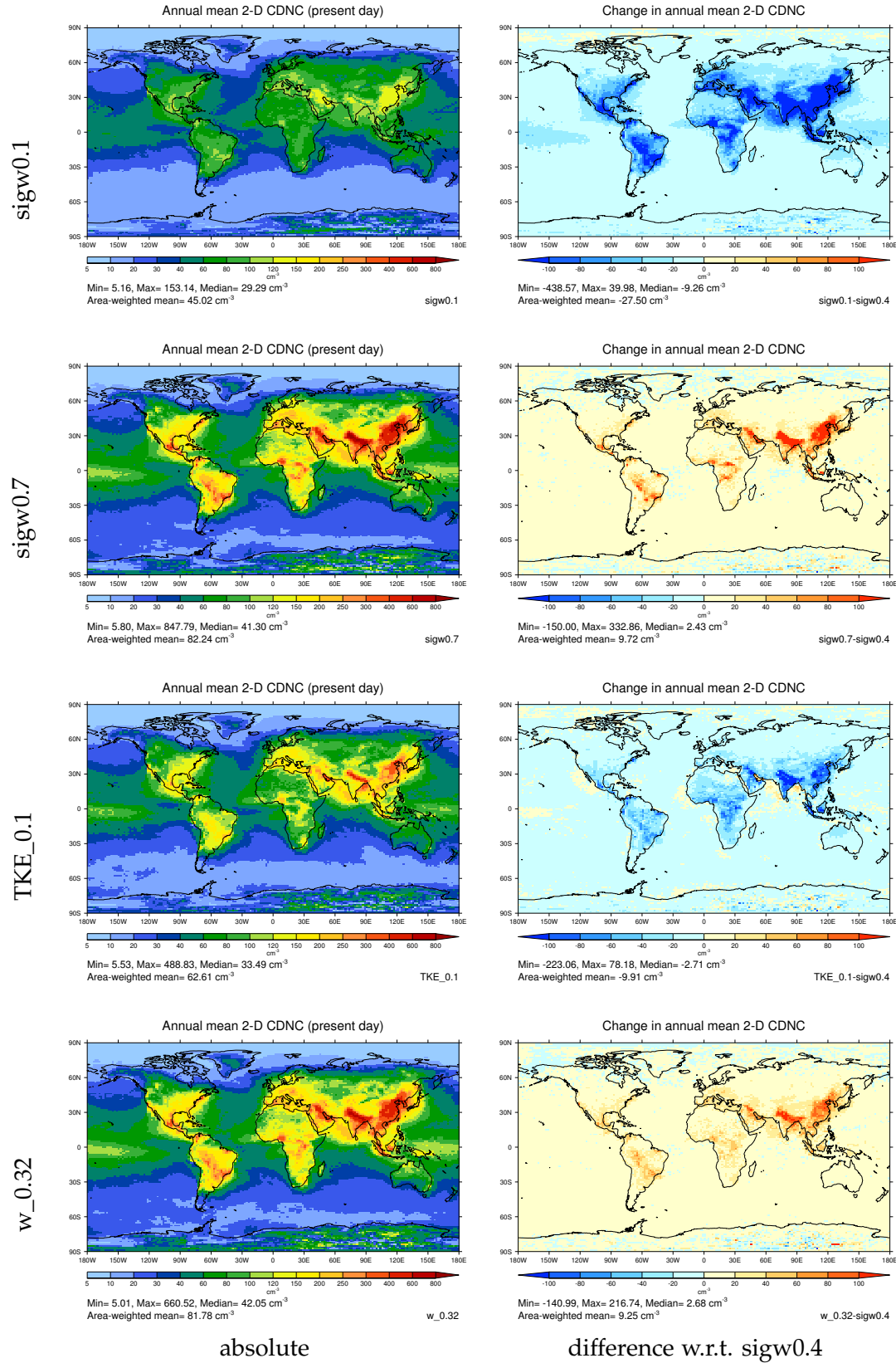


Figure 4.5: Annual mean in-cloud droplet number concentration at cloud top. Left column shows absolute value for each model configuration: sigw0.1, sigw0.7, TKE_0.1 and w_0.32 (rows). Right column shows difference of each of these configurations with respect to sigw0.4. Area-weighted means are given in the caption beneath each figure.

CDNC (a change of -9.91 cm^{-3}), and negative changes cover most of the globe, indicating regions where, on average, σ_w derived from TKE was less than 0.4 m s^{-1} . As stated previously, the effect of a lower σ_w on CDNC is particularly evident in regions of high CCN. Interestingly, there are also several regions of positive change (shown in yellow), indicating regions where the annual mean TKE-derived σ_w must have exceeded 0.4 m s^{-1} , notably in the stratocumulus region off the west coast of California, to the West of Africa and in the Bay of Bengal. The relationship between σ_w and CDNC in TKE_0.1 is explored further in Sec. 4.4.

The fourth row of Fig. 4.5 shows that using the mean updraught, w_+ , of the sigw0.4 pdf, rather than the pdf itself, leads to a general increase in CDNC which is again stronger in regions of high CCN. The global mean difference of $+9.25 \text{ cm}^{-3}$ represents a 13 % increase compared to sigw0.4, which is similar to the 10 % overestimate reported by Morales and Nenes (2010). Further differences between a model with a characteristic updraught and one with a pdf of updraught may result from the difference in variance of the CDNC over the time period between the two runs, which cannot be appreciated by comparing annual means alone, but may have consequences which emerge in the radiative effects (see Chapter 6).

Cloud droplet effective radius

Fig. 4.6 shows liquid cloud droplet effective radius, r_e , sampled at the liquid cloud-top (as described in Sec. 3.6, this is defined as the first vertical level in a column to contain liquid cloud water, starting at the top of the atmosphere in each column and working downwards. This gives a two-dimensional sample from the three-dimensional cloud field). As defined in Eq. (1.38), $r_e \propto N_d^{-\frac{1}{3}}$, so the effects of changes to σ_w will be less obvious on r_e than on N_d (especially on contour plots with a linear colour bar, as in Fig. 4.6). However, sizeable differences are still evident. (Unsurprisingly, geographic location of outstanding features are well-correlated with those of N_d in Fig. 4.5.) Decreasing σ_w from 0.4 to 0.1 m s^{-1} leads to a large global mean increase in effective radius of $1.24 \mu\text{m}$, roughly a 10 % increase compared with sigw0.4 (the direct causality is an upshot of the fact that cloud liquid water content, q_c , is determined independently by the cloud scheme in isolation from UKCA-Activate, since CDNC is diagnostic and autoconversion can only affect the next time-step). The reduction in N_d (due to this imposed decrease of σ_w) by the mechanism described above, propagates via equation Eq. (1.38) to a corresponding increase in r_e (albeit potentially attenuated by any simultaneous changes to q_c).

The non-linearity in the response of N_d to increasing σ_w described previously is also evident in the response of r_e . Increasing σ_w from 0.4 to 0.7 m s^{-1} leads to a decrease of $0.21 \mu\text{m}$ in the global mean, roughly one sixth of the change brought about by decreasing from 0.4 to 0.1 m s^{-1} .

The impacts of these changes on the annual mean net downwards shortwave radiation flux at the top of the atmosphere are a global mean change of $+2.57 \text{ W m}^{-2}$ due to a decrease of σ_w from 0.4 to 0.1 m s^{-1} , and a global mean change of -0.55 W m^{-2} due to an increase of σ_w from 0.4 to 0.7 m s^{-1} . Note that these quantities relate to the model in an un-tuned state and with present-day aerosols only. Non-linearity of the effects discussed so far indicate that it is important not to jump to conclusions regarding anthropogenic aerosol effects from differences between simulations of present-day aerosol alone. The difference in radiative flux perturbation for each model configuration due to anthropogenic aerosol will be addressed in Chapter 6.

The third row of figures in Fig. 4.6 shows the effects of using TKE-derived σ_w on r_e

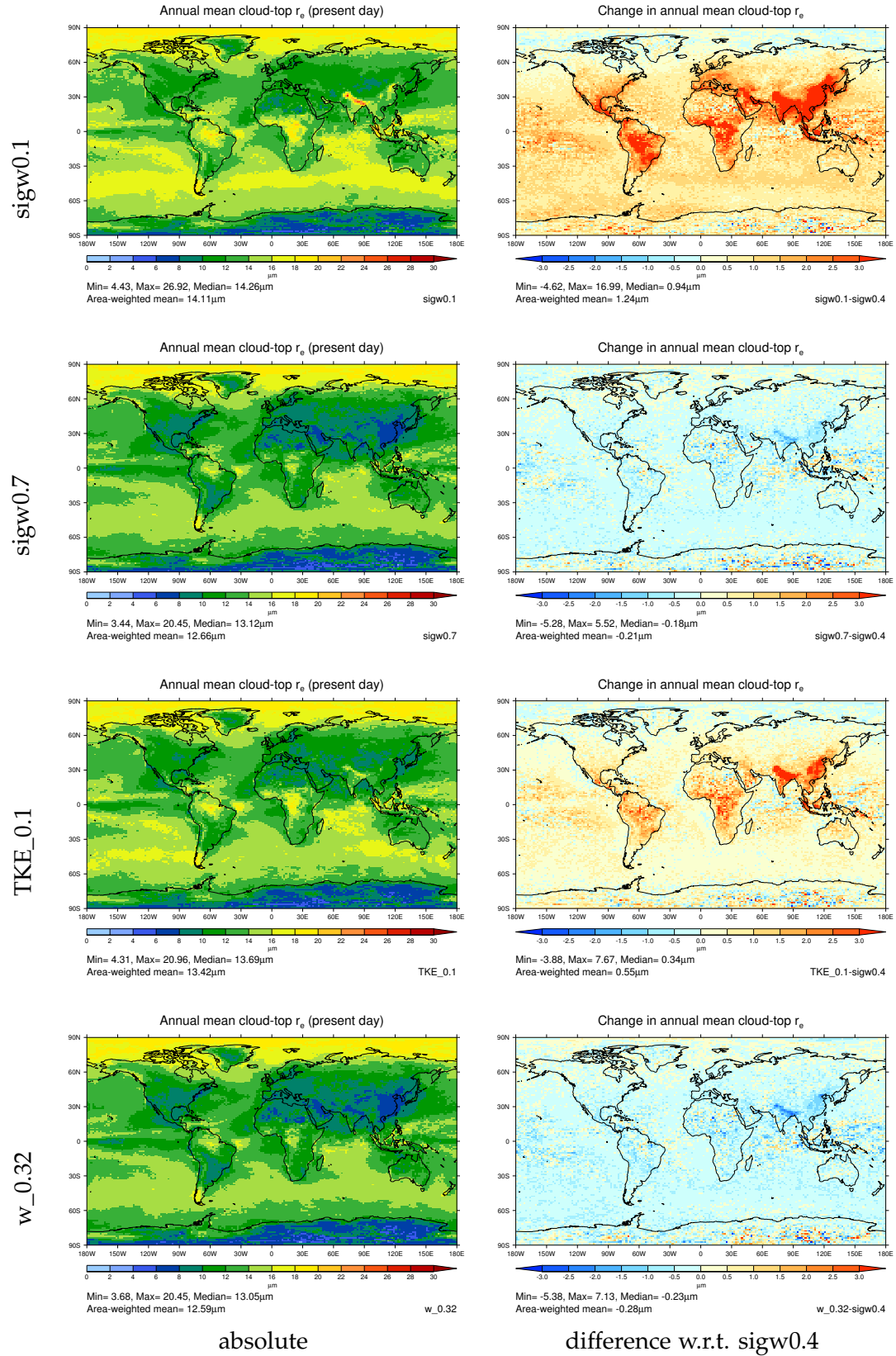


Figure 4.6: Annual mean liquid cloud droplet effective radius at cloud-top. Left column shows absolute value for each model configuration. Right column shows difference of each of these configurations with respect to sigw0.4. Area-weighted means are given in the caption beneath each figure.

(TKE_0.1 compared to sigw0.4). As mentioned before, features in the difference plot for r_e are aligned with those in the difference plot for CDNC. The increases in the stratocumulus regions in the CDNC noted above lead to less pronounced decreases in the plot of r_e , due to this inverse cube-root dependence, although they are still apparent. The most pronounced features of the third row difference plot are the increases to r_e , which occur in locations where TKE_0.1 must have $\sigma_w < 0.4 \text{ m s}^{-1}$, in regions of high CCN concentrations (e.g. over China and the Ganges plain).

Finally, the difference plot in the fourth row of figures shows a broad decrease in r_e in w_0.32 compared to sigw0.4, due to the universal increase in CDNC brought about by this relatively high fixed updraught.

Liquid water path

Fig. 4.7 shows annual mean liquid water path for each of the model configurations in Table 4.2. In the right-hand column, the difference plots of each of these configurations compared to sigw0.4 indicate that changing the variance of $P(w)$ can have an effect on the LWP. Broadly speaking, increasing σ_w corresponds to an increase of LWP, particularly over ocean, although the signal is noisy, due to differences in feedbacks between the model runs (shown in the difference plots for sigw0.1 and sigw0.7 compared to sigw0.4).

As discussed in the previous sections, increasing σ_w can lead to an increase in N_d , as shown in Fig. 4.5, because more of the smaller aerosols can activate. This in turn leads to a reduction in r_e , as shown in Fig. 4.6. These smaller droplets take longer to grow to raindrop size by the collision/coalescence process, thus decreasing the precipitation efficiency of the cloud and increasing the cloud liquid water content. This effect manifests itself in the positive differences seen when LWP(sigw0.4) is subtracted from LWP(sigw0.7) (Fig. 4.7, second row, RH column) and the negative differences seen when LWP(sigw0.4) is subtracted from LWP(sigw0.1) (Fig. 4.7, first row, RH column).

Note that this is the same mechanism that allows the cloud lifetime effect to manifest in the GCM. The difference is that here, the increase in N_d is due to an increase in σ_w between the two model runs, whereas for the classic cloud lifetime effect the increase in N_d occurs in regions where the number of CCN has increased between runs with pre-industrial and present-day aerosol emissions.

The difference plot in the third row shows a decrease in LWP between TKE_0.1 and sigw0.4, in most regions where TKE-derived $\sigma_w < 0.4 \text{ m s}^{-1}$, which is strongest in the region of highest CCN concentration over China. There is perhaps less to see in the few regions where TKE-derived $\sigma_w > 0.4 \text{ m s}^{-1}$, although this is not a clear signal. Finally, the fourth row shows that the increased CDNC in the w_0.32 run compared to sigw0.4 leads to a slight increase in LWP compared to sigw0.4, which would fit with the same mechanism described above.

4.4 Analysis of manifestation of TKE-derived σ_w

This section presents a more detailed analysis of σ_w derived from the model turbulent kinetic energy in configuration TKE_0.1. These results are highly specific to HadGEM-UKCA, but are included here because they elucidate some of the behaviour of this model configuration which applies to the comparison with data from flight campaigns shown in the next chapter.

As described in Sec. 4.2.3, σ_w in model configuration TKE_0.1 is derived from the modelled turbulent kinetic energy, where this is greater than a minimum value of 0.1 m s^{-1} .

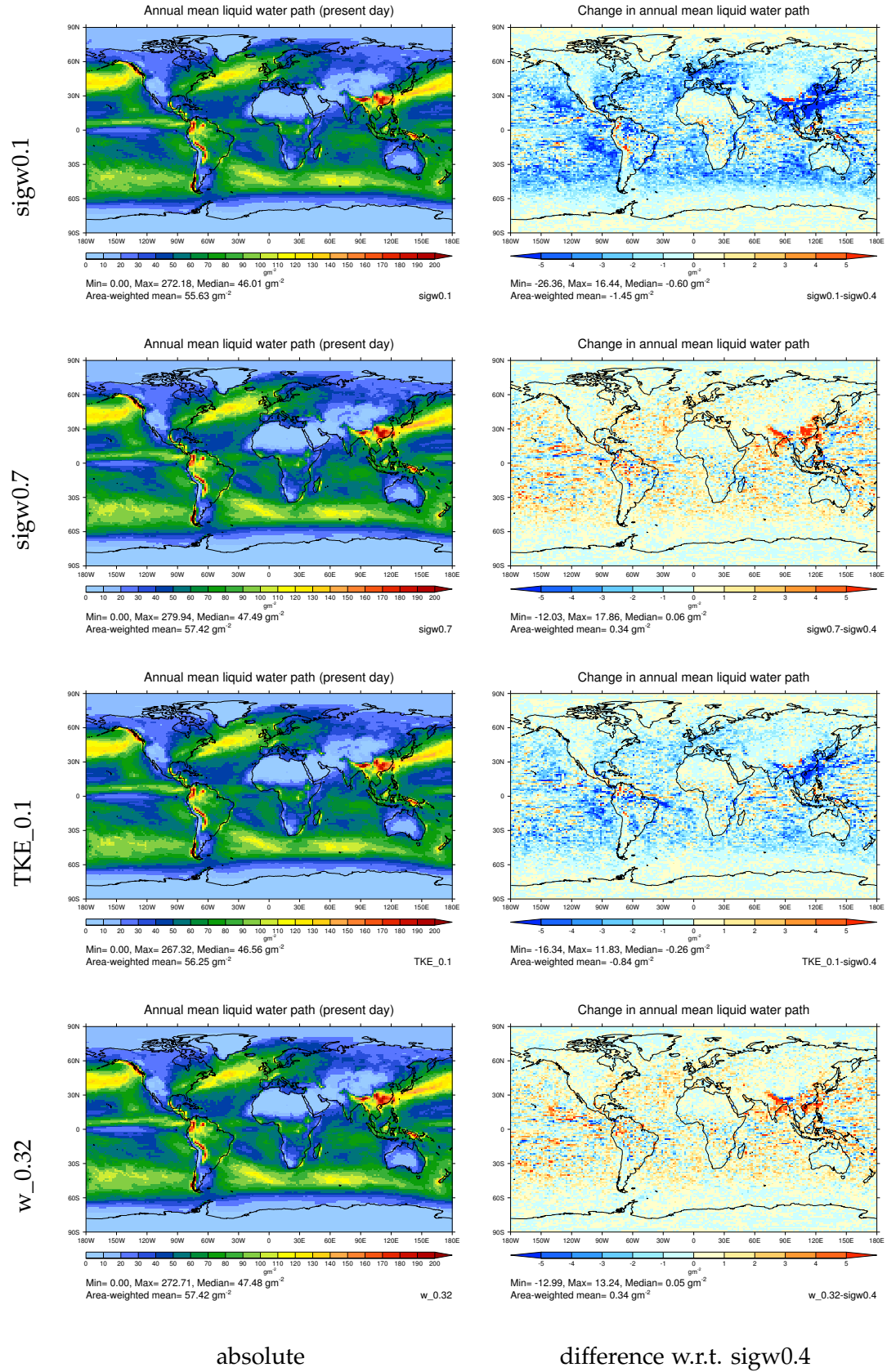


Figure 4.7: Annual mean liquid water path. LH column shows absolute value for each model configuration. RH column shows difference of each of these configurations with respect to sigw0.4. Area-weighted means are given in the caption beneath each figure.

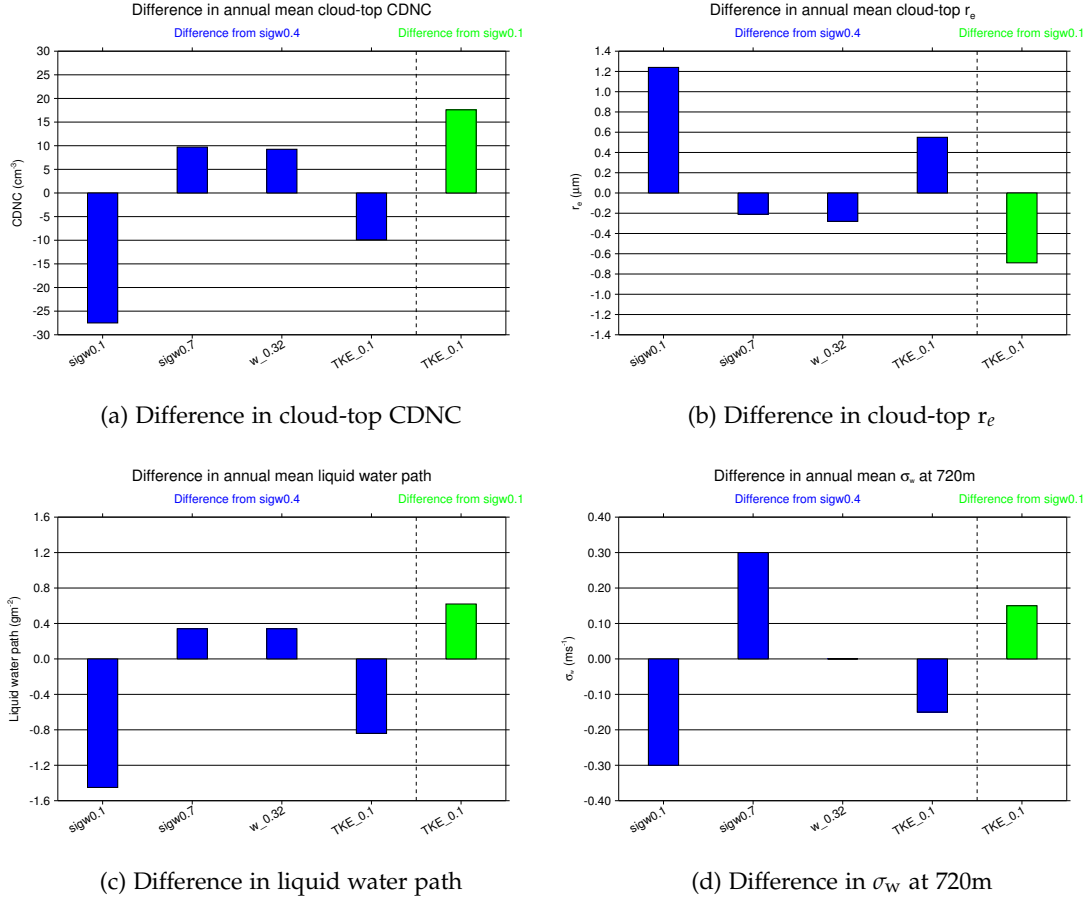


Figure 4.8: Differences in annual global area-weighted mean compared to sigw0.4 for (a) CDNC at cloud-top, (b) r_e at cloud-top, (c) liquid water path and (d) σ_w at 720m. In each chart, bars from left to right show differences of sigw0.1, sigw0.7, w_0.32 and TKE_0.1 from sigw0.4 (blue). The right-most bar in each chart shows the difference of TKE_0.1 from sigw0.1 (green).

To examine how often this minimum value is used in practice, we present histograms of σ_w at cloud-base from statistics drawn from time series of three-hourly instantaneous global fields, from the month of June (2008). This analysis was repeated for the month of December, and yields similar results (not shown).

Fig. 4.9 (a) shows a histogram of σ_w from all grid-boxes at cloud base, throughout the spatial domain of the model. Grid-boxes are defined as being at cloud-base if the grid-box is cloudy, with no cloud in the grid-box below (where cloudy is defined in the model as grid-boxes with both liquid water content, $q_c > 0$ and liquid cloud fraction, $f_c > 0$). Data in this histogram are from a month-long time series of three-hourly instantaneous fields through June 2008 (i.e. 240 time-slices). The minimum value (of 0.1 m s^{-1}) occurs just over 60 % of the time. [The maximum value of $\sigma_w = 1.83 \text{ m s}^{-1}$ is the result of the maximum value of $\text{TKE} = 5 \text{ m}^2 \text{ s}^{-2}$, imposed in the boundary layer scheme to restrict the values of TKE to an assumed suitable maximum range, being used in Eq. (4.10).]

Fig. 4.9(b) shows the same as (a), but over the lowest eleven levels only. (As described in Table 4.2, $\sigma_w = 0.1 \text{ m s}^{-1}$ in levels 12 and upwards.) The two graphs are very similar,

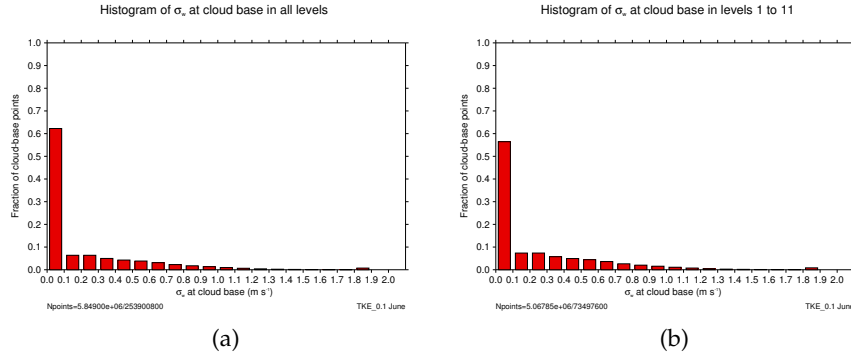


Figure 4.9: Histogram of σ_w at cloud-base from time series of three-hourly instantaneous fields for June 2008 for (a) all vertical levels, (b) levels 1 to 11 only, for model configuration TKE_0.1.

because cloud-base is nearly always within the lowest eleven model levels [as will be shown later in Fig. 4.11(a)]. Within the lowest eleven levels, the minimum value of σ_w is applied under two circumstances: either because model TKE is defined, but is lower than this minimum value; or in the case that cloud-base occurs above the maximum height of turbulent mixing within the model, such that TKE is not defined. (Within the model, turbulent kinetic energy can exist as a diagnostic throughout levels one to eleven, although its magnitude drops off rapidly above the model-defined boundary layer depth. TKE is necessarily zero above the maximum height of turbulent mixing, which can exceed the strict boundary layer depth in cases of decoupling.)

These data are split down further in Fig. 4.10, which shows histograms of σ_w at cloud-base for each of the lowest twelve model levels. In general, TKE has the greatest effect on σ_w in lower model levels and the frequency of occurrence of the minimum value increases with height.

To indicate the relative significance of the distribution of σ_w at each model level, shown in Fig. 4.10, Fig. 4.11 (a) shows the relative frequency of cloud-base within each level. The distribution is peaked, with a maximum in level 6 (hybrid height of 720 m). It is also clear that cloud-base rarely occurs in levels 12 and above, and hence the choice of the fixed value of σ_w in these levels should not have a significant impact on the final calculations of aerosol radiative effects.

Fig. 4.11 (b) and (c) illustrate how often cloud-base height, z_{CB} , occurs above the maximum height of turbulent mixing, z_{TM} . Fig. 4.11(b) illustrates that for more than half of cloud-base points, the maximum height of turbulent mixing occurs below the cloud-base level in that column, i.e. $z_{TM} < z_{CB}$. The two-dimensional histogram in Fig. 4.11(c) illustrates the vertical spatial distribution of these differences. A bi-modal distribution is evident, as expected from the presence of positive and negative differences in (b). One peak is visible beneath the 1:1 line, indicating the difference in heights for those points in which cloud base is within the scope of the turbulent mixing, i.e. $z_{CB} < z_{TM}$.

A second peak exists above the 1:1 line; showing that z_{CB} is mainly $> z_{TM}$ when $z_{TM} < 1$ km. However, since turbulent mixing is confined to the lowest eleven model levels, corresponding to a maximum hybrid height of 2880 m, and z_{TM} is mainly < 1 km, this is unsurprising.

The impacts of the vertical distribution of turbulence on σ_w in a global field is shown

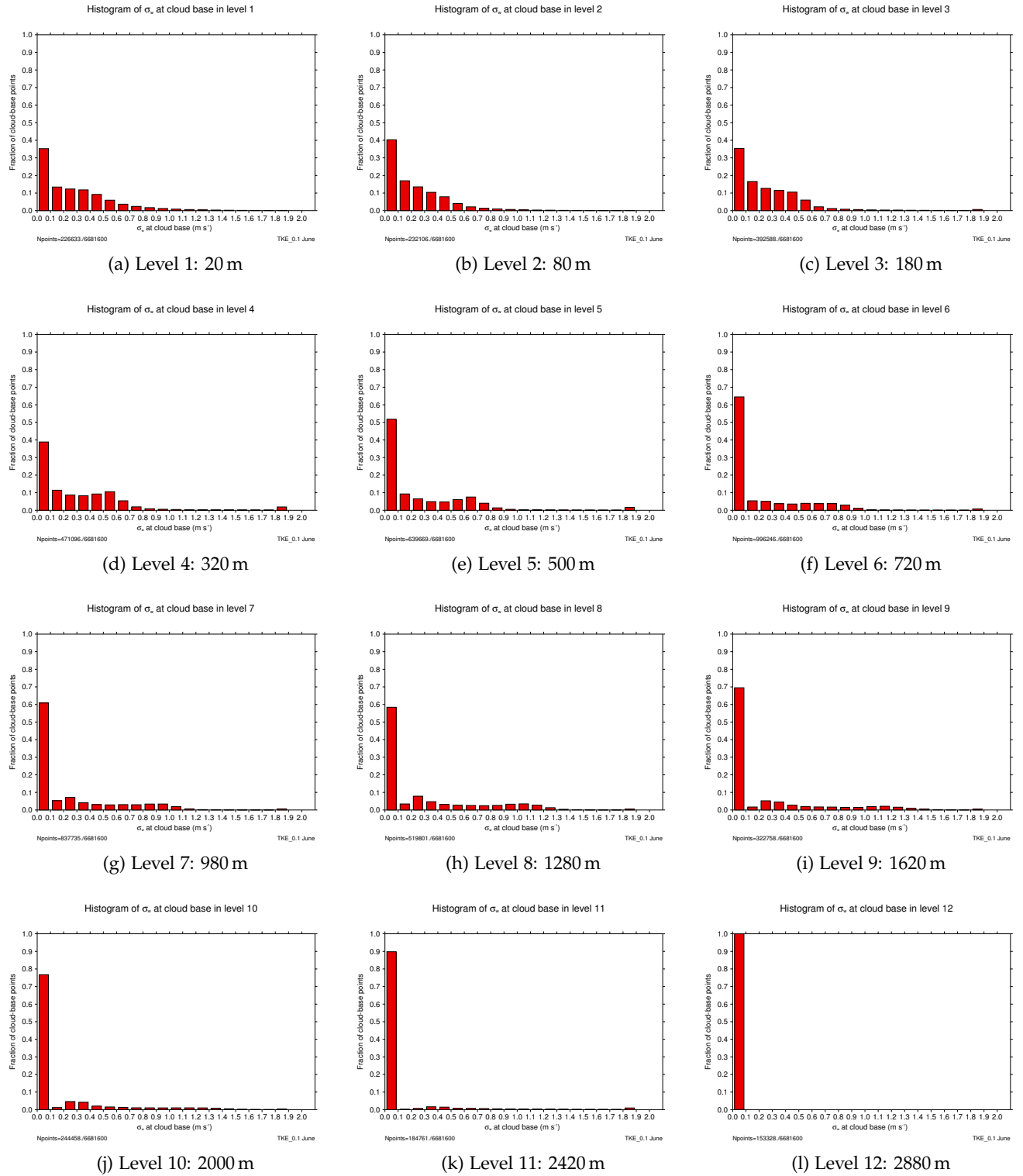


Figure 4.10: Histogram of σ_w at cloud-base from time series of three-hourly instantaneous fields from June 2008 in each model level from (a) level 1, to (l) level 12, for model configuration TKE_0.1. Hybrid height of each level is given in the caption beneath each figure. Note that in levels 12 to 38, $\sigma_w = 0.1 \text{ m s}^{-1}$ throughout.

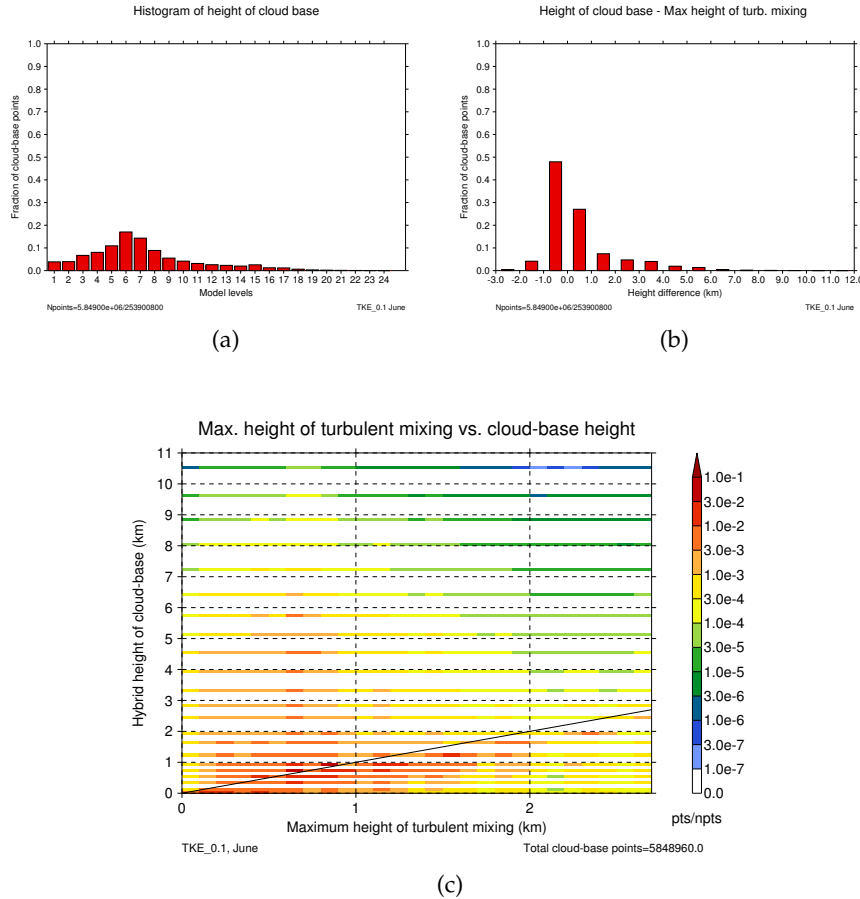


Figure 4.11: (a) Distribution of cloud-base points across model levels. (b) Distribution of height difference between cloud-base height and maximum height of turbulent mixing. Negative values indicate by how much the cloud base was below the maximum height of turbulent mixing and vice versa. (c) 2D histogram of maximum height of turbulent mixing (km) versus cloud-base height (hybrid height of levels in km). Colour indicates fraction of total number of cloud-base points. Solid diagonal line indicates 1:1. All data (a-c) from time series of three-hourly instantaneous fields points at cloud-base from June 2008 for model configuration TKE_0.1.

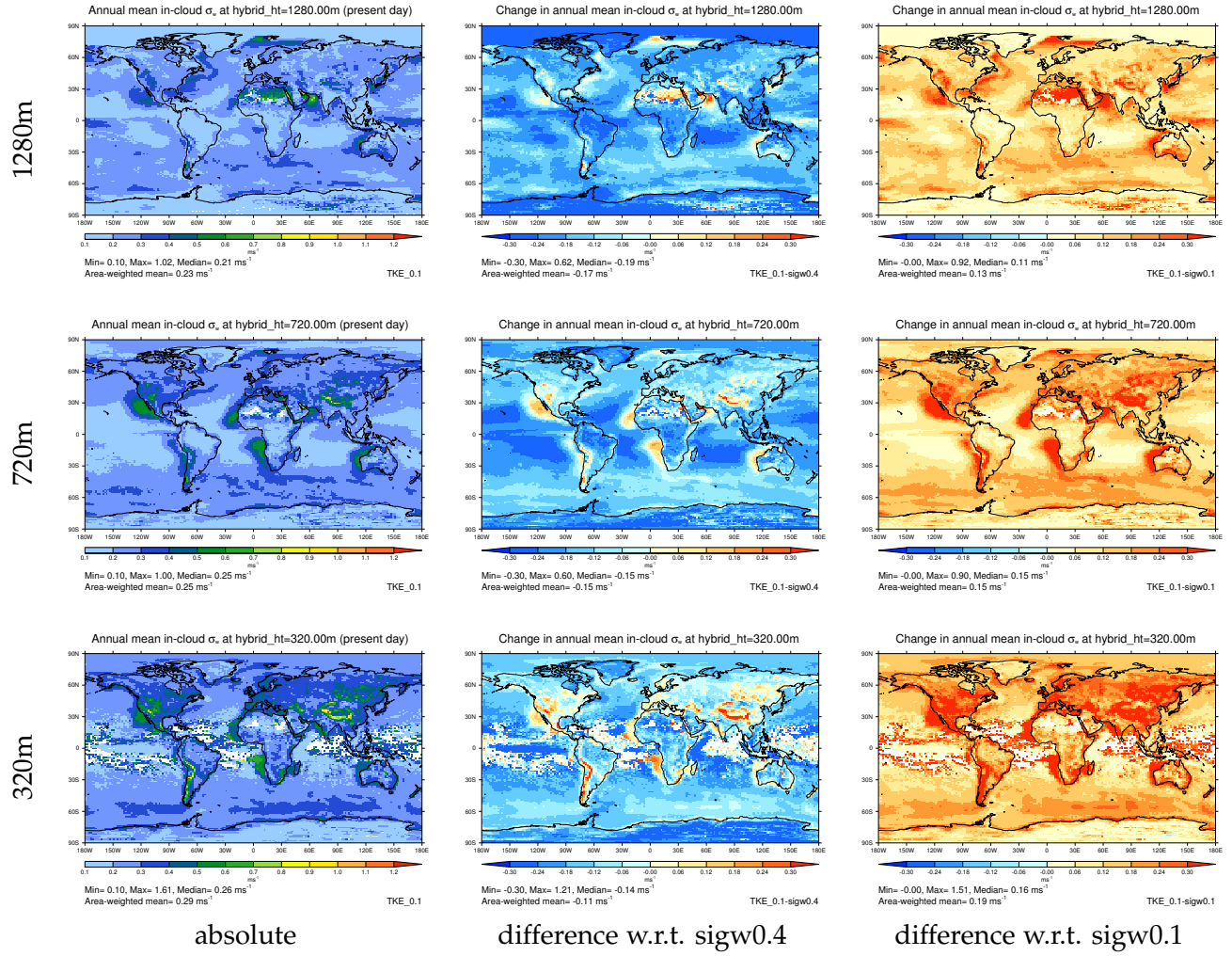


Figure 4.12: Annual mean in-cloud σ_w at 1280 m, 720 m and 320 m (model levels 8, 6 and 4) shown in rows. LH column shows configuration TKE_0.1. Central column shows difference of TKE_0.1 with respect to sigw0.4. RH column shows difference of TKE_0.1 with respect to sigw0.1.

in Fig. 4.12. The left-hand column shows annual mean in-cloud σ_w for model levels 4, 6 and 8, corresponding to hybrid heights 320 m, 720 m and 1280 m. (Note that these are annual means of in-cloud σ_w , not just σ_w at cloud-base, in line with the assumption stated in Chapter 3, that N_d throughout vertically-adjacent cloudy grid-boxes is equal to the number of aerosols activated at cloud-base. White regions indicate missing data, i.e. no cloud at any point during the year, and hence no in-cloud diagnostics.) The central column shows difference between the value of in-cloud σ_w from TKE_0.1 compared to the fixed σ_w reference case, sigw0.4 (i.e. values plotted in the central column are simply the values of TKE-derived σ_w plotted on the left-hand column, minus 0.4 m s^{-1} .) Warm colours indicate regions where TKE-derived σ_w exceeds 0.4 m s^{-1} , and cold colours indicate regions where TKE-derived σ_w falls below this value at each level. Similarly, the right hand column shows the same values of TKE-derived σ_w plotted in the left-hand column, but this time with 0.1 m s^{-1} subtracted in order to highlight areas where TKE-derived σ_w exceeds this

imposed minimum value.

Absolute values in the left hand column show that both the spatial extent and intensity of the impact of turbulent kinetic energy on σ_w decrease with height. Note that in-cloud annual means record the mean value of σ_w over all the time-steps when that grid-point was cloudy, not over all time, i.e. high values of σ_w at 1280 m over the Sahara indicate that, of the times when there was cloud over the Sahara, the TKE was also quite high, but it is not possible to infer anything about the frequency of occurrence of cloud at each grid-box (although in the case of the Sahara it is likely that this frequency was low!).

At 320 m (bottom row), σ_w exceeds the minimum value over nearly all of the land and much of the ocean. High values of 0.8 to 1.0 m s^{-1} (up to a maximum of 1.6 m s^{-1}) are visible over significant orographic features such as the Himalayas and Andes. Medium values of 0.5 to 0.7 m s^{-1} exist over parts of China, Russia and North America. Similar values also occur off the west coasts of several continents: North America, South America, Australia and two regions of Africa (to the west of Cape Verde; off the Skeleton Coast of south-west Africa).

At 720 m, the most common level for cloud-base, the most prominent regions where σ_w exceeds 0.1 m s^{-1} are the stratocumulus regions off the continental west coasts mentioned above. It is interesting to see from the middle column that at this level, TKE-derived σ_w makes significant departures from the reference case ($\sigma_w = 0.4 \text{ m s}^{-1}$) in both positive and negative directions.

The impacts of the vertical distribution of σ_w in TKE_0.1 on in-cloud droplet number concentration are shown in Fig. 4.13. Annual mean values of in-cloud N_d at each of the three vertical levels are shown in the left-hand column. In the central column, the difference in annual mean in-cloud N_d between TKE_0.1 and sigw0.4 is shown at each level. The right-hand column shows the difference in annual mean in-cloud N_d between TKE_0.1 and sigw0.1. Across all three levels, N_d from TKE_0.1 is generally lower than from sigw0.4, and differences correspond well to areas where TKE-derived $\sigma_w < 0.4 \text{ m s}^{-1}$. Regions that show reductions in N_d compared to sigw0.4 but increases in N_d compared to sigw0.1 indicate places where the value of N_d in TKE_0.1 falls in between the high values of N_d from sigw0.4 and the low values seen with sigw0.1, because $0.1 < \sigma_w(\text{TKE}) < 0.4 \text{ m s}^{-1}$. This occurs in regions of high CCN concentration, where the number of aerosols that can activate with $\sigma_w = 0.1 \text{ m s}^{-1}$ is far below the number that can activate when σ_w is increased to 0.4 m s^{-1} .

There are a few regions where N_d from TKE_0.1 just exceeds that from sigw0.4, e.g. at 320 m and 720 m in the stratocumulus region off the coast of California and two regions off the west coast of Africa, differences of up to 20 cm^{-3} are shown. These regions coincide with those regions of high σ_w over ocean in Fig. 4.12, noted previously.

However, most interestingly, other than these rather small differences, comparing the central columns of Fig. 4.12 and Fig. 4.13 demonstrates that N_d from TKE_0.1 is not greater than to N_d obtained in sigw0.4 in any of the other regions where the TKE-derived σ_w exceeded 0.4 m s^{-1} , such as the persistent stratocumulus region in the southeast Pacific. This indicates that the CDNC in such regions is limited by the low CCN concentration, as discussed in Chapter 3.

In the right-hand column, negative values indicating a reduction in N_d in TKE_0.1 with respect to sigw0.1 are likely to be due to slightly differing meteorology in the two runs (i.e. differences in the number of in-cloud points contributing to the annual mean). These points typically occur in regions of very low frequency of liquid cloud occurrence, over high latitudes at all three levels, and also for some tropical points at the 320m level, where slight differences in occurrence frequency can be significant.

Positive values in the right hand column indicate an increase in N_d from TKE_0.1

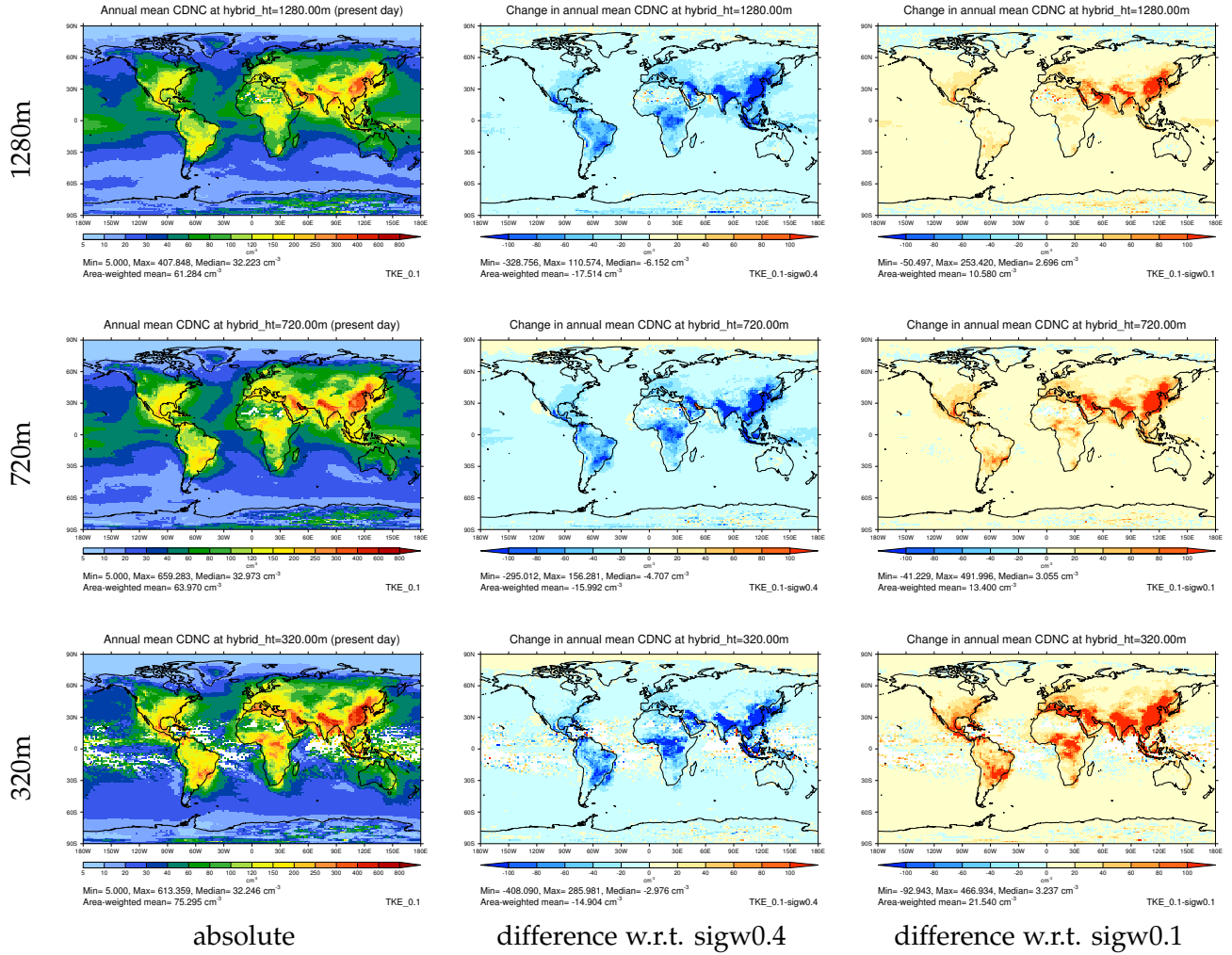


Figure 4.13: Annual mean in-cloud droplet number concentration at 1280 m, 720 m and 320 m (model levels 8, 6 and 4) in rows from top-down. LH column shows configuration TKE_0.1. Central column shows difference of TKE_0.1 with respect to sigw0.4. RH column shows difference of TKE_0.1 with respect to sigw0.1.

compared to sigw0.1 and indeed a comparison with σ_w in Fig. 4.12 shows that in the annual mean these points are well-correlated with regions where σ_w exceeds the minimum value. The prevalence of these high values decreases with height, as does the influence of TKE on σ_w .

Fig. 4.14 shows the ultimate effects of the value of TKE-derived σ_w exceeding the minimum value, on the radiatively significant cloud-top properties of N_d and r_e , and on LWP, since these plots show the differences of TKE_0.1 with respect to sigw0.1. (a) is simply a repeat of σ_w at 720 m from TKE_0.1 to give a visual reminder of the areas of high TKE in the most common cloud-base level. (b) shows number concentration of CCN at $S = 0.4\%$. The greatest increases to N_d (and decreases to r_e) occur in the regions of highest CCN number concentration, e.g. over China and the Ganges Plain, in which σ_w also exceeds the minimum value. The effect in these regions can be contrasted with Southern India, over which the CCN concentration is just as high, but the difference in σ_w smaller, and the

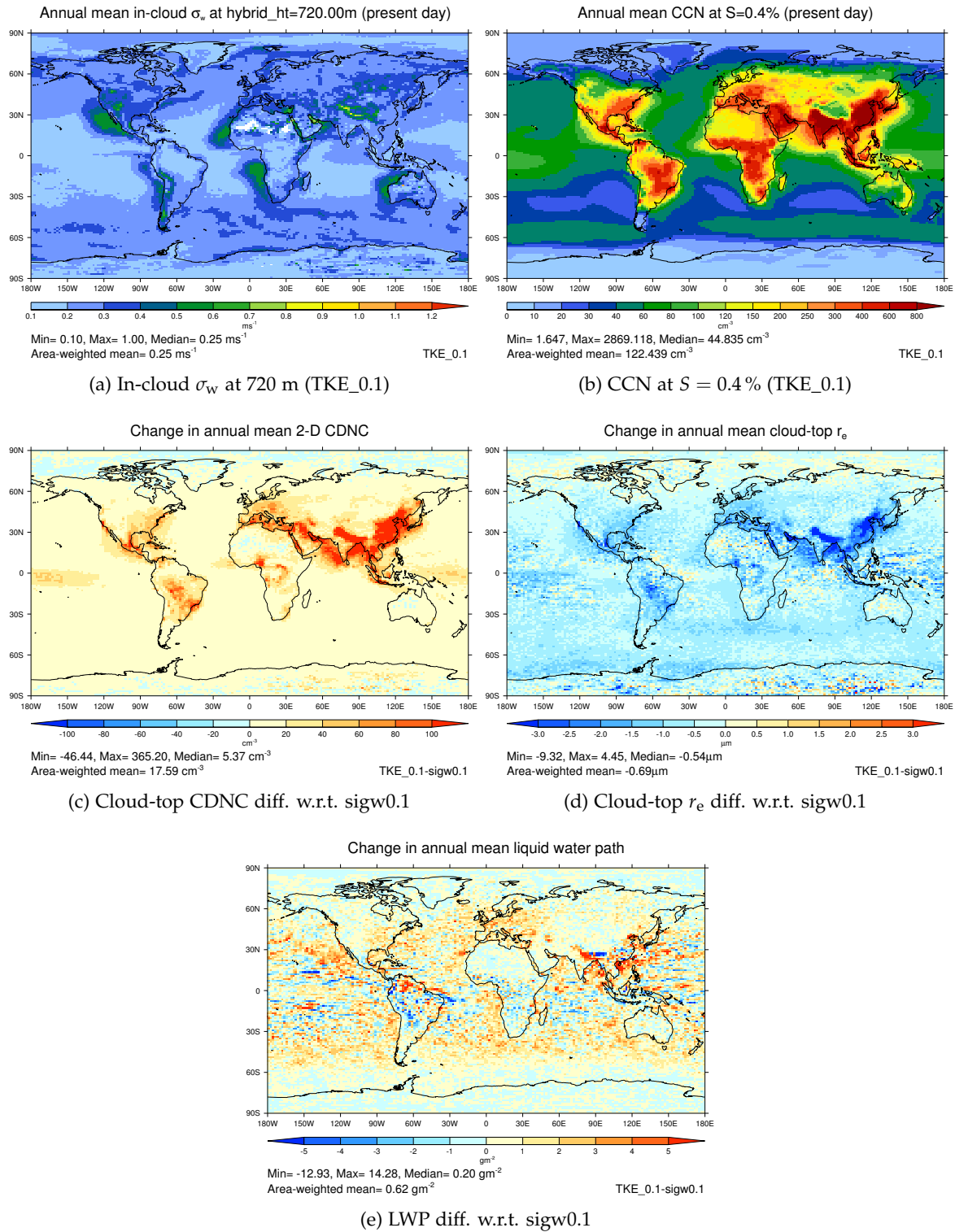


Figure 4.14: Figures (a) and (b) show annual means from model TKE_0.1 of (a) in-cloud σ_w at 720 m and (b) CCN at $S = 0.4\%$ (as described in Sec. 3.5). Figures (c) to (e) show differences between model TKE_0.1 and sigw0.1 for (c) cloud-top CDNC, (d) cloud-top r_e and (e) LWP.

increase in N_d likewise smaller.

Other land regions that show smaller increases to N_d (and decreases to r_e) are over North America and Europe. Over ocean, similar-sized increases are seen off the west coasts of California and Africa (near Cape Verde). A common feature of each of these four regions is that they all have elevated levels of σ_w in regions of medium to high CCN.

There is not such a great increase in N_d off the coast of Chile and south-west Africa, despite the similarly elevated levels of turbulence in these regions. A comparison with (b) indicates that this is potentially due to low values of CCN in these regions. This indicates that both strong updraughts and the presence of high CCN concentrations are required for high N_d . Further work is needed to establish which has the most significant effect.

Finally, Fig. 4.14(e) shows the difference in LWP between TKE_0.1 and sigw0.1. It is not so easy to correlate these differences with spatial features of either σ_w or CCN, but it is apparent that there is a broad increase in LWP between TKE_0.1 and sigw0.1, due to the increased CDNC, particularly in areas of high CCN and perhaps more prominently in regions of elevated σ_w , e.g. off the west coasts of Africa and California.

4.5 Conclusions

This chapter has presented the results of running HadGEM-UKCA with several different representations of the sub-grid-scale variability of vertical velocity. The concept of characteristic updraught velocity was addressed in box-model and GCM studies, and found to be inadequate for use across the range of aerosol conditions encountered within a GCM. Consequently, the pdf-based approach was chosen for the baseline configuration of UKCA-Activate. Despite the non-uniform nature of real world distributions of vertical velocities, and the different forms of pdf that have been (sparsely) observed in different cloud regimes, a Gaussian pdf of updraughts is assumed, and the work has focused on the parameterisation of the standard deviation of this distribution, σ_w . The effects of varying this parameter across the range of observed values was explored, as well as the possibility of deriving σ_w from the turbulent kinetic energy in the model. Annual mean fields of cloud droplet number concentration, effective radius and liquid water path are shown for each of these model configurations, as well as the results of a model run with a single fixed vertical velocity of 0.32 m s^{-1} , for comparison. Differences of each configuration from the reference case (with the median observed fixed value of $\sigma_w = 0.4 \text{ m s}^{-1}$) are shown alongside.

Cloud droplet number concentration is found to (usually, and sub-linearly) increase with increasing σ_w , and these increases are particularly strong in regions of high CCN concentration. (Consequently, cloud droplet effective radius decreases, due to the inverse relationship between the two.) In general, annual mean liquid water path is found to increase (highly non-linearly) with increasing σ_w . These changes to radiatively-significant parameters indicate that the choice of σ_w within a model may have a noticeable effect on estimates of the indirect aerosol effects. This will be quantified in Chapter 6.

Evaluation of models against *in situ* measurements from flight campaigns

5.1 Overview of flight campaigns

For the purposes of model evaluation, a range of flight campaigns that published *in situ* measurements of cloud droplet number concentration and updraught velocity statistics (in particular, cloud-base σ_w) in tabulated form, have been selected from the literature and the data collated into a database. A brief summary of the purpose and location of each campaign follows. Flight campaigns that focused on boundary layer stratiform cloud are summarised first, in Table 5.1 (NARE, ACE-2, CLOPAP, MASE, MASE-II, CSTRIFE, ACE-1, SOCEX, VOCALS-REx). The second group of campaigns sampled clouds that were more likely to be dominated by convection than large-scale ascent (RICO, PASE, CRYSTAL-FACE and ICARTT, which sampled both cumulus and stratocumulus cloud). These are summarised in Table 5.2. A brief overview of the meteorological conditions sampled by each of these campaigns is included below. Detailed descriptions of each campaign, the aircraft flown, instrumentation onboard and discussions of the implications of the scientific findings can be found in the references listed in Table 5.1 and Table 5.2.

NARE

Leaitch et al. (1996) present airborne observations from 14 flights in marine stratus over the Gulf of Maine and the Bay of Fundy during the North Atlantic Regional Experiment (NARE) in August–September 1993. These clouds formed in warm air over cooler water, and occasionally covered an underlying layer of fog. Although the main anthropogenic plume from North America was not a direct input to these clouds, some of the pre-cloud aerosol was of continental origin, and overall the region is influenced by air masses with widely varying aerosol and trace gas loading.

ACE-2

The second Aerosol Characterisation Experiment (ACE-2) featured a sub-project (CLOUDY-COLUMN) specifically dedicated to an observational study of the aerosol indirect effect in marine stratocumulus clouds, conducted over the eastern Atlantic, near the Canary Islands,

Campaign	Dates	Location	Cloud type	# Obs	Campaign focus & notes	References
NARE	08–09/1993	NW Atlantic, Gulf of Maine & Bay of Fundy	Marine St	20	Indirect aerosol effects in smooth-air, light-turbulence conditions	Leaich et al. (1996); Peng et al. (2005); Gul-tepe and Isaac (1996)
ACE-2	07/1997	NE Atlantic, near Canary Islands	Marine Sc	8	Indirect aerosol effects	Guibert et al. (2003); Pawlowska and Brenguier (2000)
CLOPAP	30/09/2005	Thames Estuary & English Channel	Marine St & Sc	4	Effect of urban aerosols on cloud microphysics	Romakkaniemi et al. (2009)
MASE	07/2005	NE Pacific, off California	Marine Sc	9	Aerosol–cloud relationships	Lu et al. (2007)
MASE-II	07/2007	NE Pacific, off California	Marine Sc	11	As for MASE, plus ship tracks	Lu et al. (2009)
CSTRIPE	07/2003	NE Pacific, off California	Marine Sc	52	Coastal Stratocumulus	Meskhidze et al. (2005)
ACE-1	15/11–14/12/1995	Southern Ocean, S of Australia	Marine Sc	14	Summertime clouds in remote region	Yum et al. (1998); Bates et al. (1998); Hudson et al. (1998)
SOCEX	20/01–09/02/1995	Southern Ocean, W of Tasmania	Marine Sc	8	Cloud properties in unpolluted region. No σ_w .	Boers et al. (1998)
VOCALS-REx	16/10–15/11/2008	SE Pacific, off Chile	Marine Sc	69	Structure of Sc-topped BL and lower troposphere. (Obs split into three longitude bins.)	Bretherton et al. (2010)

Table 5.1: Details of flight campaigns through stratiform clouds used for model evaluation.

Campaign	Dates	Location	Cloud type	# Obs	Campaign focus & notes	References
RICO	12/2004– 01/2005	Antigua, NE Caribbean	Cu	14	Rain In Cumulus over Ocean	Hudson and Noble (2009); Hudson and Mishra (2007)
PASE	09/08– 07/09/2007	Mid-Pacific, near Christmas Island	Cu	27	Pacific Sulphate Expt. Very few clouds	Hudson and Noble (2009)
CRYSTAL-FACE	07/2002	Key-West, Florida	Deep convective & cirrus	20	Tropical anvils and cirrus layers. Obs of liquid cloud properties at warm cloud base	Meskhidze et al. (2005); Conant et al. (2004); VanReken et al. (2003)
ICARTT	2004	Ohio, N America	Continental St & Cu	27	Highly polluted clouds near power stations	Fountoukis et al. (2007) Zhang et al, JGR 2007

Table 5.2: Details of flight campaigns through non-stratiform clouds used for model evaluation.

in July 1997 (Brenguier et al., 2000a). Four of these flights were through clouds forming in clean marine air masses, with droplet concentrations ranging from 55 to 128 cm⁻³. The other four flights were through clouds in air masses with varying degrees of pollution, with flight-mean droplet concentrations ranging from 128 to 244 cm⁻³.

CLOPAP

The scientific aim of the Cloud Processing of Regional Air Pollution Over Land and Sea (CLOPAP) campaign was to investigate the effect of urban aerosols on the microphysics of clouds downwind of major pollution sources. Flights took place over several days, but measurements are only published from one of these days (Romakkaniemi et al., 2009), when flights were conducted over East Anglia and the Thames Estuary. Wind, predominantly from the southwest, blew polluted air masses originating from the London area into the region of study. (Aircraft and instrumentation details are available at <http://www.faam.ac.uk/public/campaigns/index.html>)

North East Pacific: MASE-I, MASE-II & CSTRIFE

The persistent stratocumulus cloud deck over the north-east Pacific, off the coast of Monterey, California, is a relatively well-studied regime, primarily due to its significant climatic importance, but also perhaps due to its close proximity to several major atmospheric research institutes (California Institute of Technology, US Naval Research Laboratory, UCLA, etc). Three recent flight campaigns have published measurements of both CDNC and updraught velocity statistics of clouds in this region and have been included in the database.

The Coastal STratocumulus Imposed Perturbation Experiment, CSTRIFE, published a table of measurements of CDNC and updraught velocity mean and standard deviations from 52 flights in July 2003, through the eastern Pacific stratocumulus region (Meskhidze et al., 2005).

The MARine Stratus/Stratocumulus Experiment took place in two phases, MASE-I in July 2005 (Lu et al., 2007), and MASE-II in July 2007 (Lu et al., 2009). July was chosen because it is the month in which stratocumulus coverage of the region is typically maximal. Both phases were designed to investigate aerosol–cloud relationships. This region of the Pacific atmosphere experiences air masses of very clean background air as well as those with a much stronger anthropogenic influence. The presence of several major shipping lanes in the region also provides a useful opportunity for studying the effects of localised aerosol perturbation on marine stratocumulus due to ship tracks.

Since both phases of MASE and CSTRIFE have sampled the same cloud regime in the same geographic region, in July (albeit of different years), these measurements will be aggregated together for the purposes of comparison with model results, and these combined observations will be referred to as NEPac-MASE-CSTRIFE.

ACE-1

The first Aerosol Characterisation Experiment, ACE-1 (Bates et al., 1998), made *in situ* measurements of summertime marine stratocumulus clouds over the Southern Ocean in a remote region, far from anthropogenic aerosol sources. Only boundary layer stratus clouds were considered in this analysis.

SOCEX

Boers et al. (1998) present six case-studies of stratocumulus clouds observed during the summer phase (between 20th January and 9th February 1995) of the Southern Ocean Cloud EXperiment (SOCEX), which investigated the microphysical and radiative properties of clouds in the unpolluted background region over the Southern Ocean. Flights were made to the west of Tasmania, where anthropogenic influences are minimal, but a clear seasonal cycle in CCN is observed, compared to the winter phase of SOCEX (Boers et al., 1996), likely due to the seasonality in the production of CCN-precursor dimethylsulphide (DMS) (Charlson et al., 1987; Gras, 1990).

For the cases presented, Boers et al. (1998) report that the atmospheric boundary layer was in various stages of decoupling, such that cumulus cloud near the top of the well-mixed surface mixed layer was covered by a uniform sheet of stratocumulus cloud at the top of the atmospheric boundary layer. The top of the surface mixed layer was often close to the mean lifting condensation level, while the layer containing the upper clouds was conditionally unstable and convective in nature.

Measured cloud droplet number concentrations varied from 50 to 150 cm⁻³, roughly three times the number observed during the winter phase of the measurement campaign (10 to 40 cm⁻³), among the lowest CDNC ever recorded (Boers et al., 1996). Updraught velocity was not reported for either phase of SOCEX.

VOCALS-REx

The VAMOS (Variability of the American MOnsoon Systems) Ocean-Cloud-Atmosphere-Land Study Regional Experiment, VOCALS-REx, was a major international field campaign to make multi-platform, co-ordinated observations of the coupled climate system of the south-east Pacific ocean (see Wood et al. 2011). The data used for this evaluation represent a very small subset of this campaign. These measurements were taken on a transect along 20°S between the coast of Northern Chile and a buoy 1500 km offshore. Measurements from individual flight legs have been combined into three longitude bins (70–75W, 75–80W and 80–85W), so the model data have been split in the same way. This transect documents the typical structure of the southeast Pacific stratocumulus-topped boundary layer, which is generally capped by a strong (10 to 12 K), sharp temperature inversion. Near to the coast, this layer is typically 1 km deep, fairly well-mixed and topped by thin non-drizzling stratocumulus with $N_d > 200 \text{ cm}^{-3}$. As it extends away from the shoreline, the boundary layer deepens (to about 1.6 km) and the vertical structure tends to become decoupled. These offshore stratocumulus clouds have strong mesoscale organisation, much higher peak liquid water paths, persistent drizzle, and lower values of $N_d < 100 \text{ cm}^{-3}$. The increased dryness of the lower free troposphere away from the influence of the Andes promotes strong cloud-top radiative cooling and stronger turbulence in these offshore clouds. More details are given by Bretherton et al. (2010).

RICO

The Rain in Cumulus over Ocean (RICO) field project was designed to investigate warm rain initiation in unpolluted cumulus clouds (Rauber et al., 2007). The island of Antigua, at the northeast corner of the Caribbean Antilles was chosen as a suitable location, due to the predominance of east-northeast winds, bringing in air from a long unpolluted stretch out over the Atlantic. The campaign focused on the effect of pre-existing or pre-cloud aerosol

on cloud microphysics, by looking at correlations between CDNC, CCN, aerosol number concentration and drizzle drop concentration.

PASE

The location of the PACific Sulfate Experiment (PASE), a clear air chemistry project, was specifically chosen for its dryness and remoteness (Christmas Island is in an equatorial dry zone). However, clouds could not be avoided on eleven of the flights and these provide observations of small, shallow, cumulus clouds in a very remote location in the mid-Pacific (Hudson and Noble, 2009).

CRYSTAL-FACE

Meskhidze et al. (2005) report a subset of an *in situ* dataset of cumuliform clouds made during NASA's Cirrus Regional Study of Tropical Anvils and cirrus Layers–Florida Area Cirrus Experiment (CRYSTAL-FACE), near Key-West, Florida, in July 2002 (VanReken et al., 2003; Conant et al., 2004). The main aim of this project was to investigate the physical properties and formation processes tropical cirrus cloud. Tropical cirrus anvils are generated by strong, deep convective systems, and the evolution of these systems was a major focus of CRYSTAL-FACE. One aircraft was dedicated to characterising the sub-cloud aerosol layer feeding the convective system above, as well as measuring liquid cloud properties at the cloud base to aid understanding of warm cloud microphysics. It is these measurements that have been added to the database.

ICARTT

The 2004 International Consortium for Atmospheric Research on Transport and Transformation experiment (ICARTT) sampled 27 cumuliform and stratiform clouds. A particular feature of the dataset is the sampling of highly polluted clouds within the vicinity of power plant plumes in Cleveland and Detroit.

In-cloud average CDNC correlated well with cloud updraught velocity: clouds with $N_d > 1000 \text{ cm}^{-3}$ typically had $w > 1 \text{ m s}^{-1}$; the range $500 < N_d < 1000 \text{ cm}^{-3}$ typically had $w \sim 0.5 \text{ m s}^{-1}$; and the range $300 < N_d < 500 \text{ cm}^{-3}$ typically had $w \sim 0.25 \text{ m s}^{-1}$. High droplet concentration (even in clouds with low updraught velocity) was indicative of the high aerosol loading in almost all of the clouds profiled; average total aerosol concentration in flights just below cloud base ranged from 4200 to $13\,300 \text{ cm}^{-3}$.

5.2 Model time-series for each flight campaign

This section examines high time-resolution data from HadGEM-UKCA, co-located in time and space with each of the flight campaigns listed above. Co-location processing was applied to high time-resolution output from each of the configurations listed in Table 4.2, but in the discussion in this section, the focus is on the behaviour of configuration TKE_0.1. Note that the time series shown in this section only include data from the model, and that comparisons with measurements from each campaign are reserved until Sec. 5.3.

As described in Sec. 4.2, HadGEM-UKCA was run for 12 months (following a three-month spin-up period) in the configurations listed in Table 4.2 that represent sub-grid-scale variability of vertical velocity with a Gaussian pdf. The configurations differ in the value of the variance of this pdf, which takes on three fixed values, spanning the range of data from

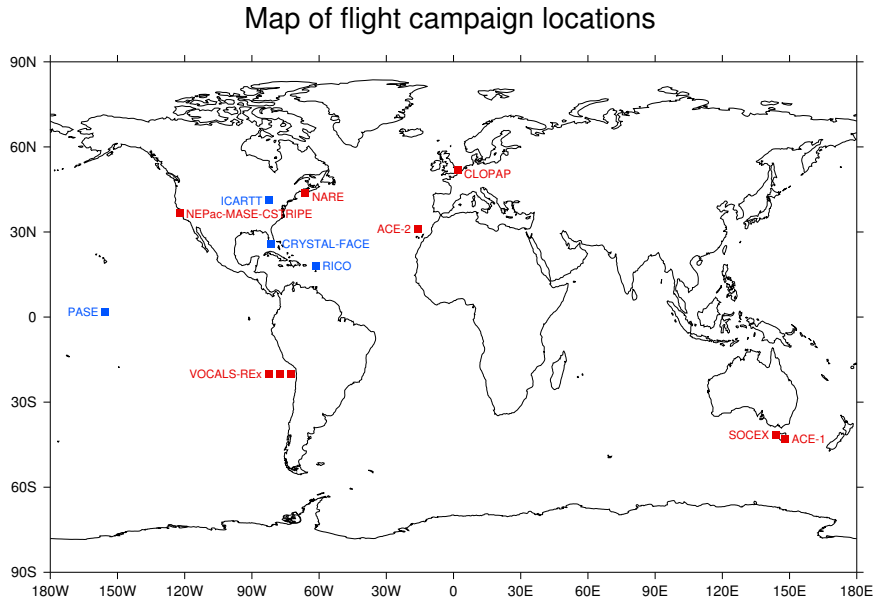


Figure 5.1: Map of flight campaign locations. Marine stratiform flight campaigns are labelled in red. Campaigns through other types of cloud are shown in blue.

all of the flight campaigns considered in this chapter, and one configuration (TKE_0.1) in which the variance of the pdf is derived from the turbulent kinetic energy in the model.

For all simulations, meteorology was nudged to ECMWF re-analysis fields (as described in Sec. 3.1) for the year 2008, chosen notionally to include the time period of the most recent flight campaign in the database, VOCALS-REx (October–November 2008). In addition to the monthly means, annual means and daily satellite-sampled diagnostics required for the model evaluation in Chapter 3, the following diagnostics were output at three-hourly intervals: in-cloud droplet number concentration (N_d), standard deviation of in-cloud updraught velocity distribution (σ_w), boundary layer depth (z_{BL}), maximum height of turbulent mixing (z_T), and CCN at seven of the most commonly-measured supersaturations from the dataset used for CCN evaluation in Sec. 3.5. For each flight campaign in the database, model output was co-located with the campaign measurements in time and space as follows.

Time

Model output was taken from the whole month of the simulation corresponding to the month of the mean date of the measurements, since flight campaigns had a typical duration of three to four weeks. Model diagnostics were output instantaneously every three hours, but only one of these three-hourly time-slices was used per day for the appropriate month at each location, since campaigns typically made one flight per day (albeit sometimes sampling more than one cloud). For campaigns that published the time that each flight started, model output for each day of the month was sampled at the time-slice closest to the mean flight start time. For campaigns that did not publish times of flights, the time-slice closest to noon local time was sampled, since most flights were made during daylight hours. (In the case of

PASE, for which two of the flights were at night, the model output is split by day/night in the analysis shown in Fig. 5.16.)

Space

The geographic extent of most of the flight campaigns was comparable in area to the horizontal model grid spacing ($1.875^\circ \times 1.25^\circ$), so model output for each campaign was sampled for the grid-box nearest to the mean longitude and latitude values of the flights recorded. For campaigns that did not publish latitude and longitude of each flight, these have been estimated from figures of flight paths, or from supplementary mission reports.

Most of the measurements considered in the database were taken at cloud-base. In the case of multi-layer clouds, it is assumed that aircraft were flown through the lowest layer, since low-level clouds were the focus of most of the campaigns. Therefore, modelled in-cloud properties were therefore sampled at the cloud-base of the lowest cloud in a column at that time-step, the level of the first ‘cloudy’ grid-box above the surface, where ‘cloudy’ is defined as any grid-box in which both liquid water content and liquid cloud fraction are greater than zero.

5.2.1 Guide to time series figure panels

Figures 5.2 to 5.8 show month-long time series of daily instantaneous model output, sampled according to the above criteria, i.e. matched to each flight campaign by month and flight time, co-located to nearest horizontal grid-box, and sampled vertically at the lowest model cloud-base, for model configuration TKE_0.1 (see Table 4.2).

Within the panel of figures for each campaign, sub-figures (a) show time series of cloud-base height (z_{CB}), boundary layer depth (z_{BL}), and maximum height of turbulent mixing (z_T). z_{BL} and z_T are defined at every time-step, so the points are joined by a continuous line. Cloud-base, z_{CB} , can only be defined when there is liquid cloud in the selected column at each time-slice, and hence this line is not always continuous. Likewise, CDNC at cloud-base, in sub-figures (b), and in-cloud σ_w at cloud-base, in sub-figures (c), only exist when liquid cloud is defined in the column.

In the background of sub-figures (a), coloured blocks indicate the value of σ_w calculated from TKE (where this is non-zero) using $\sigma_w = \sqrt{2/3 \text{TKE}}$. Note that this σ_w is not confined to in-cloud points and no minimum value is imposed. Instead, yellow is used to signify points where TKE is non-zero, but low enough to cause σ_w calculated in this way to be less than 0.1 m s^{-1} . These blocks of colour are included to give an indication of the time evolution and variability of the vertical profile of boundary layer turbulence at each location, as relevant to the σ_w . Integers printed next to z_{BL} points indicate boundary layer type determined by the boundary layer scheme, as illustrated in Fig. 4.3.

Sub-figures (d) show time series of CCN at a range of fixed supersaturations, $S = 0.2, 0.3, 0.4, 0.5, 0.75, 0.85$ and 1.0% , calculated from aerosol properties in the lowest model level (20 m), as in Sec. 3.5.

CDNC at any individual point is dependent on the updraught velocity and CCN spectrum at each grid-point, and time series of σ_w and CCN have both been included alongside CDNC in order to give some indication of the relative significance of updraught velocity parameterisation versus modelled aerosol to the CDNC variability in each case. In general, a comparison between figures (a), (b) and (c) for each campaign shows that for points with cloud-base within the boundary layer, the magnitude of in-cloud σ_w at each point affects the value of CDNC to varying extent, tempered by the CCN concentration.

Note that, as prescribed, points with cloud-base above the boundary layer (i.e. $z_{CB} > z_T$) have $\sigma_w = 0.1 \text{ m s}^{-1}$, and CDNC generally has a low value at the corresponding point in sub-figure (c).

5.2.2 Boundary layer type, cloud base height and σ_w

As mentioned in Sec. 4.2.3 and shown in Fig. 4.3, the UM boundary layer scheme distinguishes between seven different boundary layer types. For reference, summaries of each type (as defined by Lock et al. (2000)) are repeated here:

1. Stable boundary layer, possibly with non-turbulent cloud (no cumulus, no decoupled stratocumulus, stable surface layer).
2. Stratocumulus over a stable surface layer (no cumulus, decoupled stratocumulus, stable surface layer).
3. Single mixed layer, possibly cloud-topped (no cumulus, no decoupled stratocumulus, unstable surface layer).
4. Decoupled stratocumulus not over cumulus (no cumulus, decoupled stratocumulus, unstable surface layer).
5. Decoupled stratocumulus over cumulus (cumulus, decoupled stratocumulus, unstable surface layer).
6. Cumulus-capped layer (cumulus, no decoupled stratocumulus, unstable surface layer).
7. Shear-dominated unstable layer.

Since the value of the TKE-derived σ_w at cloud-base is so dependent on whether the cloud-base is within the boundary layer (i.e. $z_{CB} < z_T$), it is useful to look at the value of σ_w in conjunction with boundary layer type at each point in the time series, as shown in sub-figures (a) for each campaign.

The first four marine stratiform campaigns have $z_{CB} < z_T$ for almost all points (NARE, ACE-2, NEPac, ACE-1). A high degree of variability of cloud-base σ_w is evident, with maximum values of σ_w up to 1.32 m s^{-1} , and the lower limit on σ_w is only hit a small number of times. The boundary layer is mainly of type 3 (single mixed layer, possibly cloud-topped), with a few exceptions. For the NE Pacific case (Fig. 5.5), the boundary layer is always type 3, and z_{CB} only exceeds z_T once. The drop in σ_w to $\sigma_w^{\min}$ at this point (July 15th) stands out clearly in corresponding sub-figure (c). The situation is similar for ACE-1, Fig. 5.6; the boundary layer is nearly always type 3, and $z_{CB} > z_T$ only twice, and again these points where $\sigma_w = \sigma_w^{\min}$ are clear in sub-figure (c). For NARE, again, $z_{CB} < z_T$ with only a couple of exceptions when the lowest cloudy level occurs above the boundary layer. The time series for ACE-2 (Fig. 5.3), shows a few cases where the stratocumulus layer becomes decoupled above a layer of cumulus (type 5). In these cases, z_T and z_{BL} separate (with z_T extending up to the top of the decoupled layer), and the TKE profile splits into two distinct layers. Unfortunately, the lowest cloudy level often seems to occur in between these two layers, where there is little modelled TKE and so, in these cases, $\sigma_w^{\min}$ is used for aerosol activation at cloud-base.

This effect is also evident in the most remote VOCALS region, VOCALS-80-85. Three longitude points along the 20S VOCALS-REx flight transect are sampled by the model (72.5W, 77.5W and 82.5W) to correspond with the 5° longitude-binned data published

by Bretherton et al. (2010). Near the coast, (VOCALS-70-75, Fig. 5.8) the boundary layer is mostly type 3 (a single mixed layer). However, moving west along the transect away from the coast, through VOCALS-75-80 (Fig. 5.9) to VOCALS-80-85 (Fig. 5.10), this single mixed layer develops into a decoupled stratocumulus layer fed by cumulus at the top of an unstable surface layer below (nearly all type 5). This evolution of boundary layer with distance away from the coast matches remarkably well to observations of boundary layer characteristics in this region documented in the literature (Bretherton et al., 2010). However, as described for ACE-2, in type 5 boundary layers, the modelled turbulence profile splits into two distinct layers, with cloud-base falling between the two where there is negligible TKE, causing the σ_w in the decoupled stratocumulus layer to be unrealistically low.

The remaining two marine stratiform case studies, SOCEX (Fig. 5.7) and CLOPAP (Fig. 5.4) show more variation in boundary layer type, and more instances when $\sigma_w = \sigma_w^{\min}$, either because the cloud base falls between the decoupled layer and the surface layer beneath (BL types 4 or 5) or above z_T and z_{BL} altogether. In the case of SOCEX, the latter occurs when the boundary layer is type 6, indicating a cumulus-topped boundary layer. In these cases, z_{CB} occurs just above z_{BL} , where there is no TKE in the model, and hence σ_w is unrealistically low.

This is a particular problem for the two tropical cases, PASE and RICO, which are dominated by type 6 boundary layers, with cloud-base nearly always occurring just above the top of the boundary layers. This happens because cloud-base has been defined simply as the lowest level in which $q_c > 0$ and cloud fraction > 0 . For these shallow cumulus cases, cloud water is detrained into the large-scale cloud scheme at the cloud-top, so the calculated cloud-base may not be at the actual physical base of convective clouds, where these extend over more than one model level. Since UKCA-Activate has only been designed to represent aerosol activation in stratiform clouds, it is no surprise that it fails in these convectively-dominated regions. Comparison with observations (shown below in Fig. 5.16) indicates that in these cases at least, $\sigma_w^{\min} = 0.1 \text{ m s}^{-1}$ is far too low a value to impose, and a better solution is required, ideally informed by the boundary layer type itself.

For the mid-latitude cases, ICARTT (Fig. 5.12) and CRYSTAL-FACE (Fig. 5.11), the boundary layer is mainly type 3 (indicating a single mixed layer, no cumulus, possible stratocumulus) with some instances of type 6 (cumulus-capped layer). However, cloud-base usually occurs in the level above z_T and so again, σ_w is restricted to the minimum value. These four campaigns demonstrate the importance of the choice of $\sigma_w^{\min}$, since this defines the width of $P(w)$ for nearly all clouds with a cloud base above the boundary layer. In fact Fig. 4.11 (b) shows that in the month analysed (June, three-hourly instantaneous fields at all grid-points), cloud base is above the maximum height of turbulent mixing $\sim 65\%$ of the time, and Fig. 4.9 (a) shows that $\sigma_w = \sigma_w^{\min} = 0.1 \text{ m s}^{-1}$ 62% of the time. This high frequency of occurrence of $\sigma_w^{\min}$ indicates that model results ought to be tested for their sensitivity to this parameter. However, due to the absence of any measurements of $\sigma_w^{\min}$ in above-boundary layer clouds needed to make a better choice of $\sigma_w^{\min}$ and the relatively transient nature of such clouds compared to the more persistent and hence more radiatively-important marine stratocumulus clouds, this area of parameterisation development has been left for future work.

5.2.3 Time series of CCN and CDNC

Of course, updraught velocity is not the only factor affecting CDNC, which is also fundamentally dependent on the abundance, size and composition of the local aerosol population. These aerosol properties can be relevantly summarised by considering the number of cloud

condensation nuclei that would form at various fixed levels of supersaturation at each point (see Sec. 3.5). Sub-figures (d) show time series of CCN at various fixed supersaturations simulated at the lowest model level in configuration TKE_0.1 (although CCN concentrations are very similar for all of the configurations considered). CCN at different supersaturations ($S = 0.2, 0.3, 0.4, 0.5, 0.75, 0.85$ and 1.0%) are shown in different colours. In several cases (ACE-2, PASE, SOCEX, all VOCALS cases) the number of CCN is not greatly affected by increasing supersaturation, which links back to similar results seen in the CCN evaluation in Sec. 3.5. This indicates a high fraction of activation, even at low supersaturation (0.2%). In these cases it is a high fraction of activation of a relatively low number of aerosols. This indicates that no matter what the value of updraught velocity at these points, the number of droplets forming is likely to be limited by the abundance of CCN. However, in other cases, [CRYSTAL-FACE, Fig. 5.11(d), ICARTT Fig. 5.12(d), NEPac-MASE-CSTRIPE, Fig. 5.5(d)], CCN has a wide spread with increasing supersaturation, indicating that the lack of suitable aerosol is unlikely to be the limiting factor in CDNC¹.

Stratiform clouds

As described above, the model simulations of the marine stratiform campaigns [ACE-1 (Fig. 5.6), ACE-2 (Fig. 5.3), NARE (Fig. 5.2), NEPac-MASE-CSTRIPE (Fig. 5.5), SOCEX (5.7), CLOPAP (Fig. 5.4) and VOCALS (Fig. 5.8, 5.9 and 5.10)], have cloud-base within the boundary layer for the majority of the time. As a result, σ_w takes on a broad range of values from 0.1 to 1.0 m s^{-1} . In these cases, the time-series of CDNC appear well-correlated with the time-series of surface CCN at each location, with any blips coinciding with the occasions when the lower limit of $\sigma_w = 0.1 \text{ m s}^{-1}$ is hit in the time-series of σ_w . The low values of CDNC for ACE-2 and SOCEX are clearly dominated by low numbers of CCN in these regions. The high values of CDNC seen for the NEPac-MASE-CSTRIPE case are a combination of high aerosol abundance and consistently high σ_w . On the one occasion in the NE Pacific case when the model cloud-base occurs above the boundary layer causing the minimum value of σ_w to be used (15th July), CDNC drops compared to the previous day (14th), despite the increasing trend in CCN.

Non-stratiform clouds

The CRYSTAL-FACE time-series (Fig. 5.11), only contains five points at cloud-base, indicating that warm clouds are not commonly produced by the model at this location (Florida) in July. As described above, CRYSTAL-FACE was designed to investigate the development of cirrus and tropical cirrus anvils, which are prevalent in the region. Although observations indicate that liquid droplets are present at the bottom of these deep convective systems, and the aircraft made measurements of at least 20 such clouds during the campaign, this frequency is not well captured by the model. Either this time-series is a poor sample of the typical behaviour of the region, or this process is partially misrepresented by the model. On the days when the model does produce liquid cloud, the base is above the boundary layer for four out of five points.

¹Physically, the value of σ_w has an effect on the number of droplets that form through its influence on the maximum supersaturation, dependent on the CCN spectrum. One way of thinking about the links between CDNC and CCN plotted in these time series is that the value of σ_w on each day in sub-figures (c) gives a rough indication of which colour of point (CCN at what supersaturation) on that day in sub-figures (d) may be used to estimate the CDNC in sub-figures (b).

The ICARTT time-series (Fig. 5.12) also contains very few points, and again, cloud-base is above the boundary layer in seven out of nine of these. In both of these examples the low imposed value of σ_w above the boundary layer restricts the number of aerosols which activate, and the CDNC is low despite the high CCN concentrations.

In contrast, both RICO and PASE show a relatively high number of points in the time-series, indicating the near-daily occurrence of some liquid cloud in each column at the sampled time, but again as described previously, the level at which liquid water from these convective clouds is detrained into the large-scale cloud mainly occurs above the boundary layer so σ_w takes on the minimum value. In RICO, this appears to restrict CDNC but in the very remote PASE region CDNC is more limited by the lack of suitable CCN.

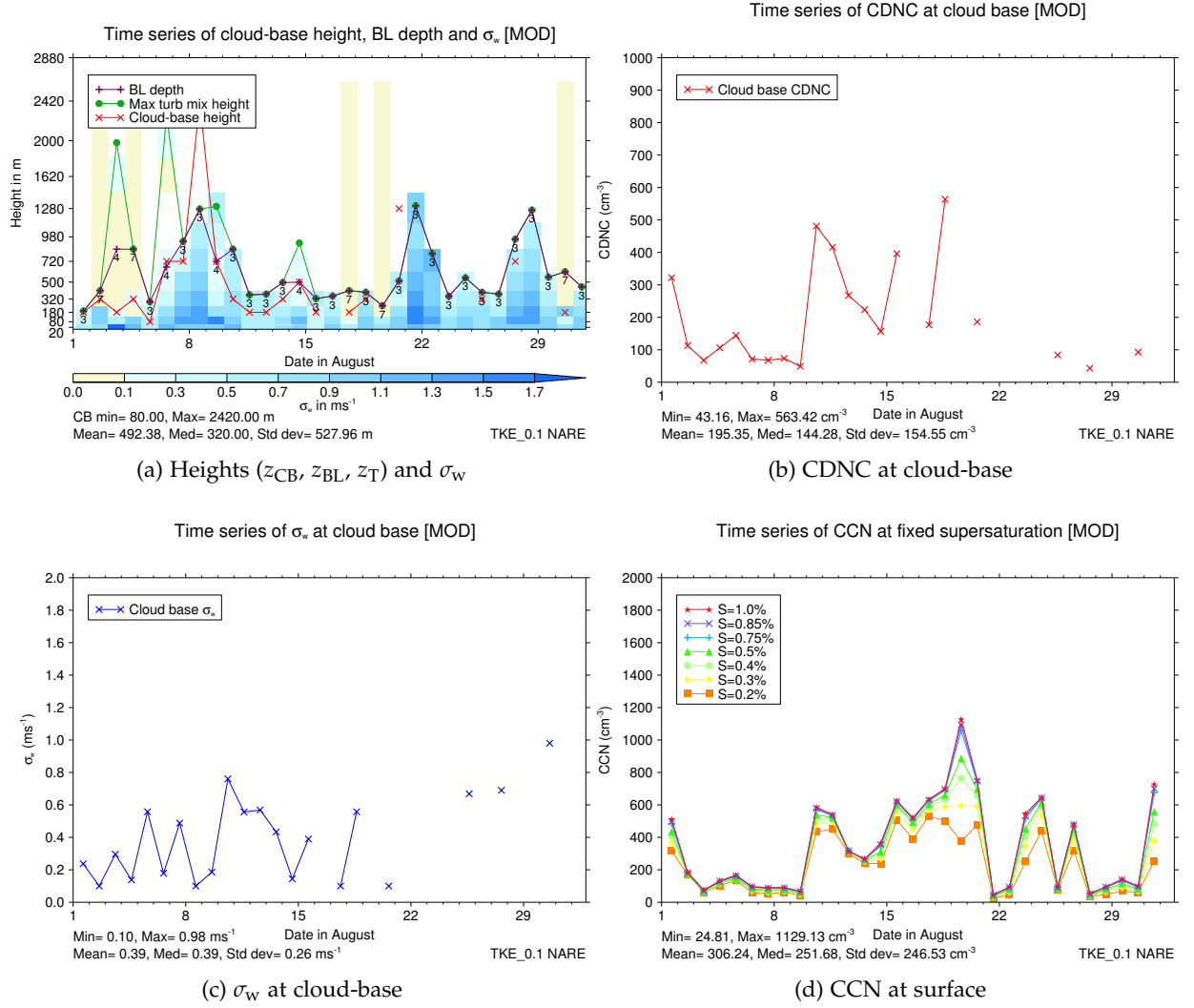


Figure 5.2: Modelled time series for experiment TKE_0.1, co-located at the NARE flight campaign location and month. (Note that only model data are shown here.) Each sub-figure shows time series of (a) cloud-base height (i.e. lowest cloudy level), z_{CB} [red crosses], boundary layer depth, z_{BL} [purple plus signs], and maximum height of turbulent mixing, z_T [filled green circles]. Coloured blocks indicate the value of $\sigma_w = \sqrt{2/3}TKE$ (where TKE is non-zero). Note that this σ_w is not confined to in-cloud points. Yellow shows where the minimum value of $\sigma_w^{\min} = 0.1 \text{ m s}^{-1}$ would be imposed. Integer labels at z_{BL} points indicate boundary layer type as illustrated in Fig. 4.3; (b) CDNC at cloud-base [red crosses]; (c) in-cloud σ_w at cloud-base [blue crosses]; (d) CCN at $S=0.2$ [orange], 0.3 [yellow], 0.4 [pale green], 0.5 [green], 0.75 [blue], 0.85 [purple], 1.0% [pink], in the lowest model level (20 m above surface).

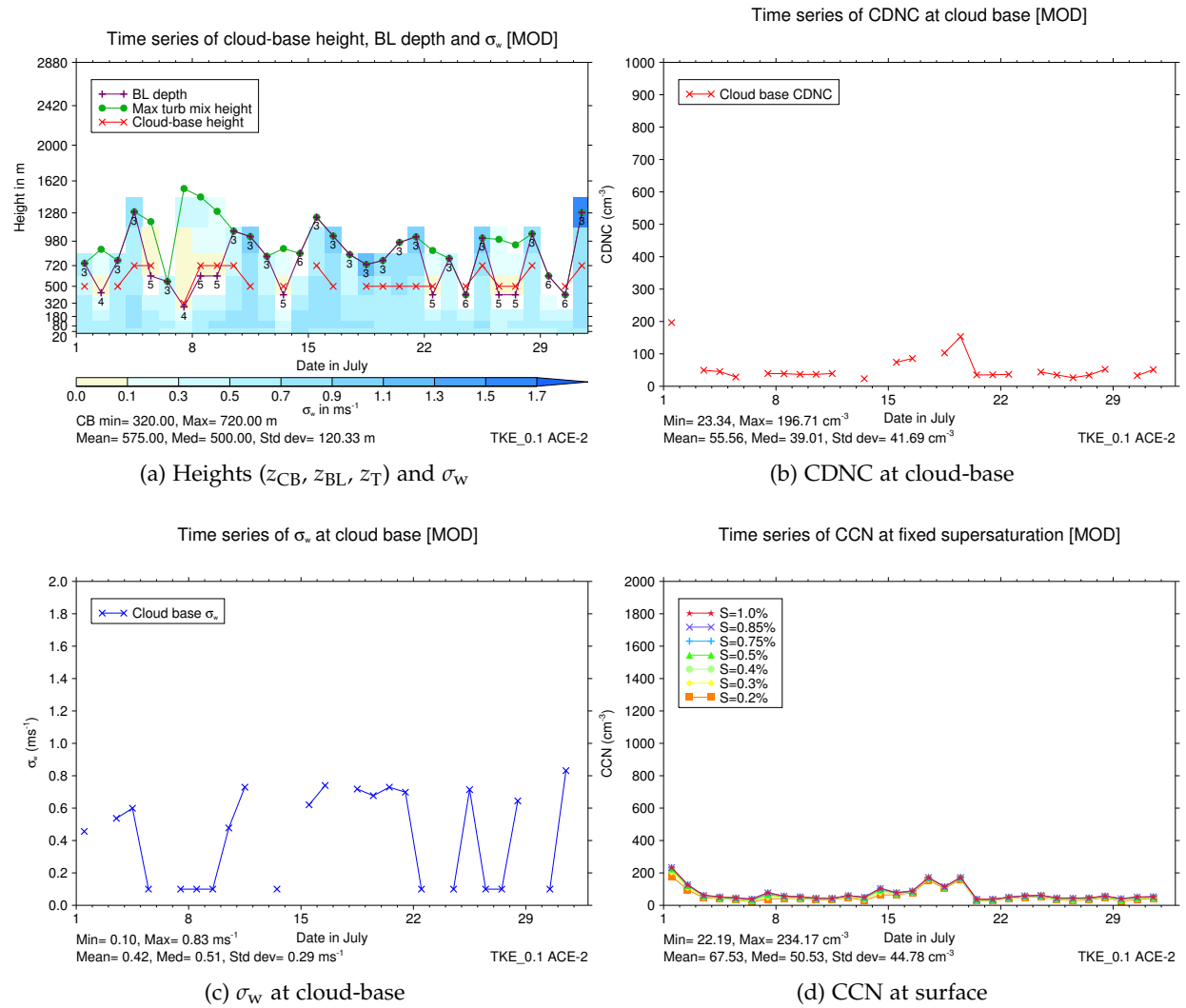


Figure 5.3: As Fig. 5.2 for model data co-located with ACE-2.

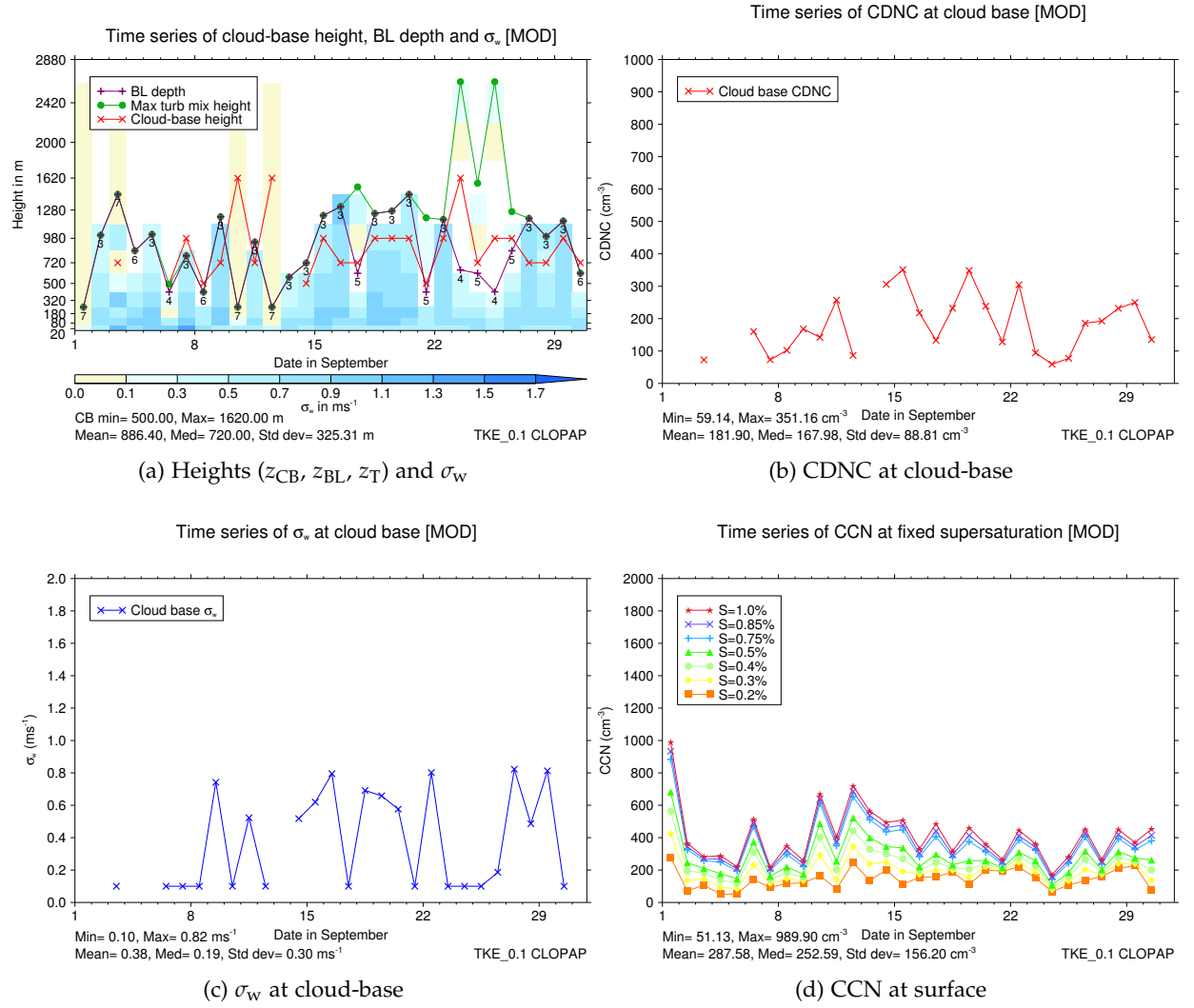


Figure 5.4: As Fig. 5.2 for model data co-located with CLOPAP.

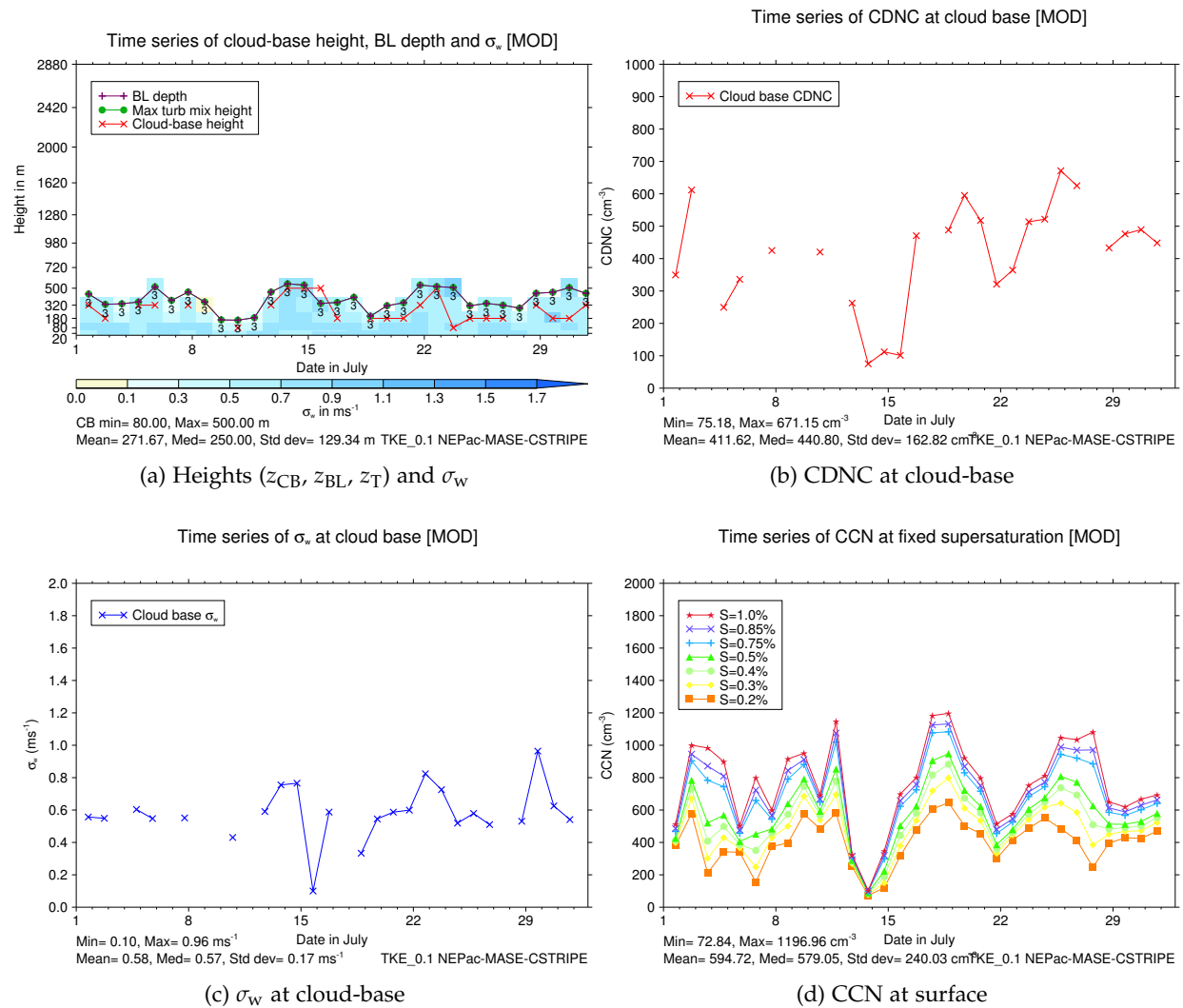


Figure 5.5: As Fig. 5.2 for model data co-located with NEPac-MASE-CSTRIPE.

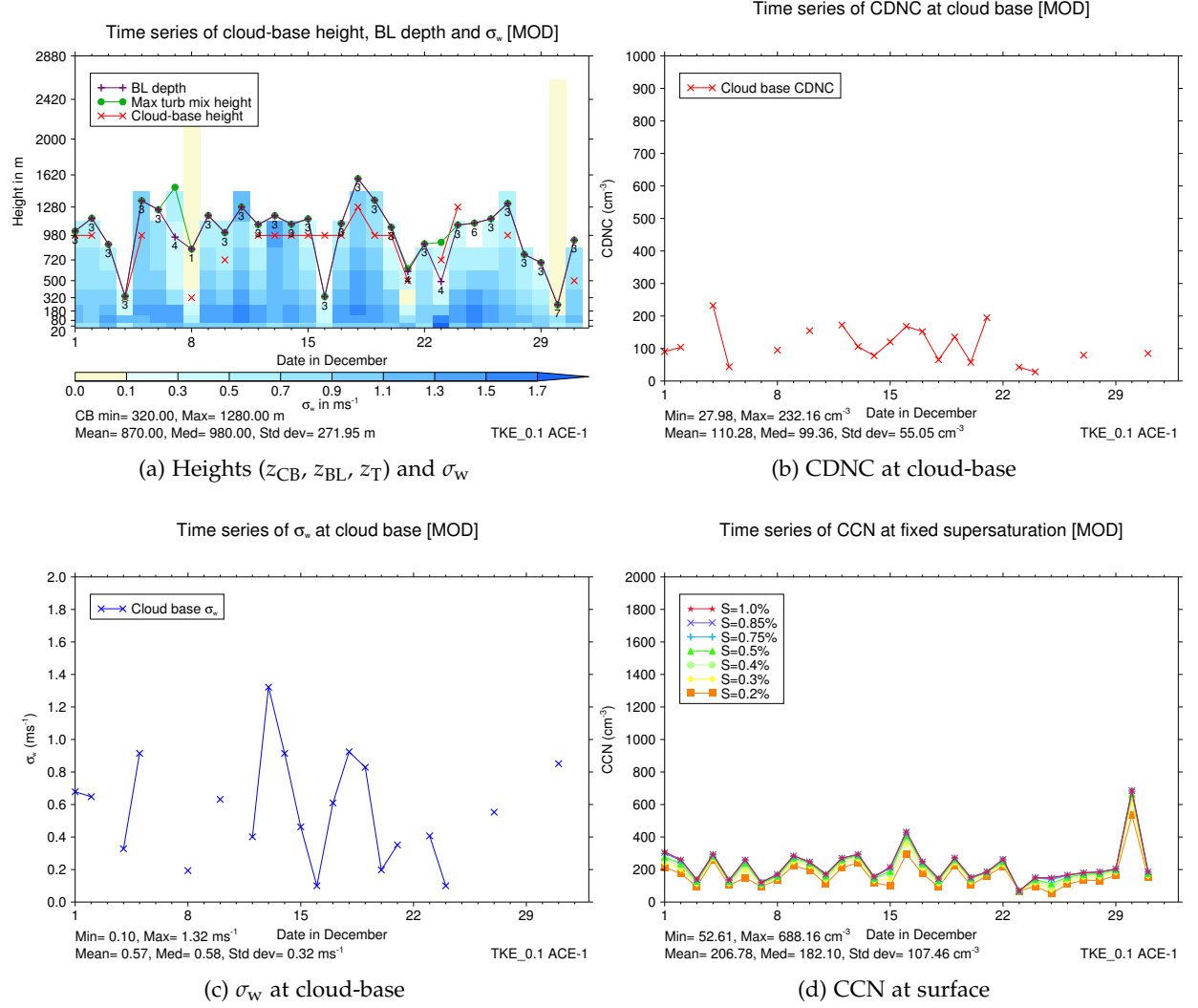


Figure 5.6: As Fig. 5.2 for model data co-located with ACE-1.

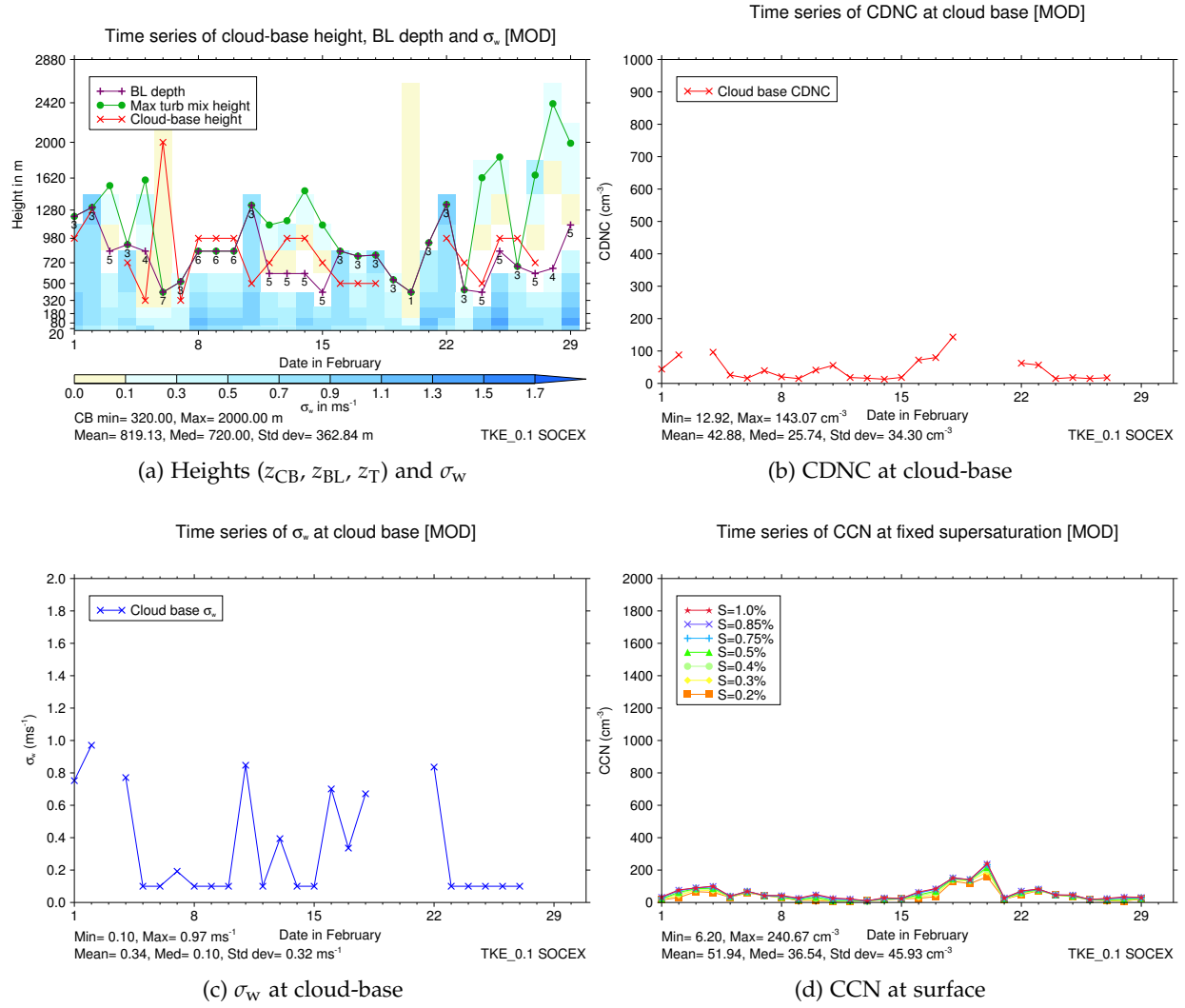


Figure 5.7: As Fig. 5.2 for model data co-located with SOCEX.

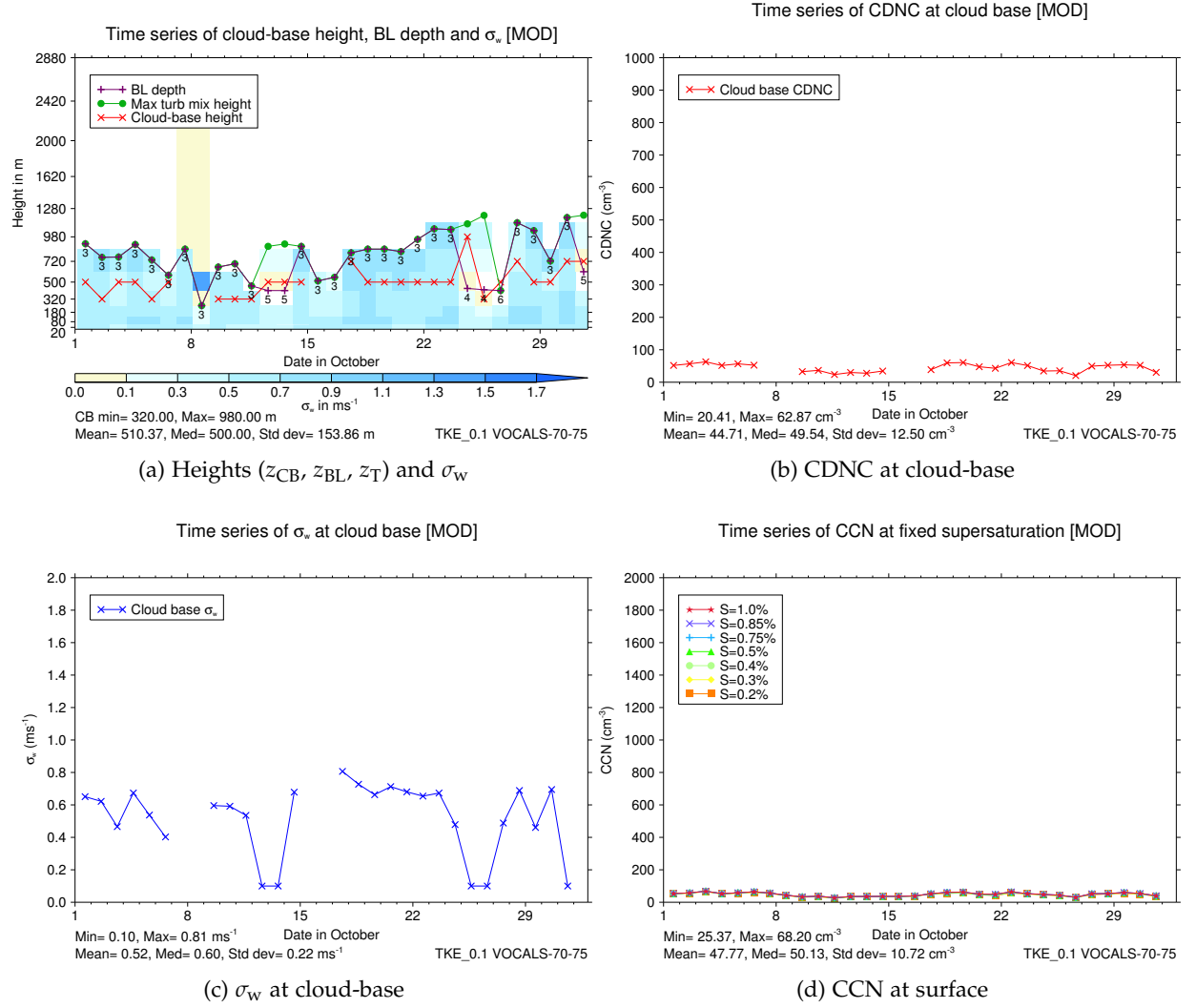


Figure 5.8: As Fig. 5.2 for model data co-located with VOCALS-70-75, i.e. the model is sampled at 72.5W, 20S.

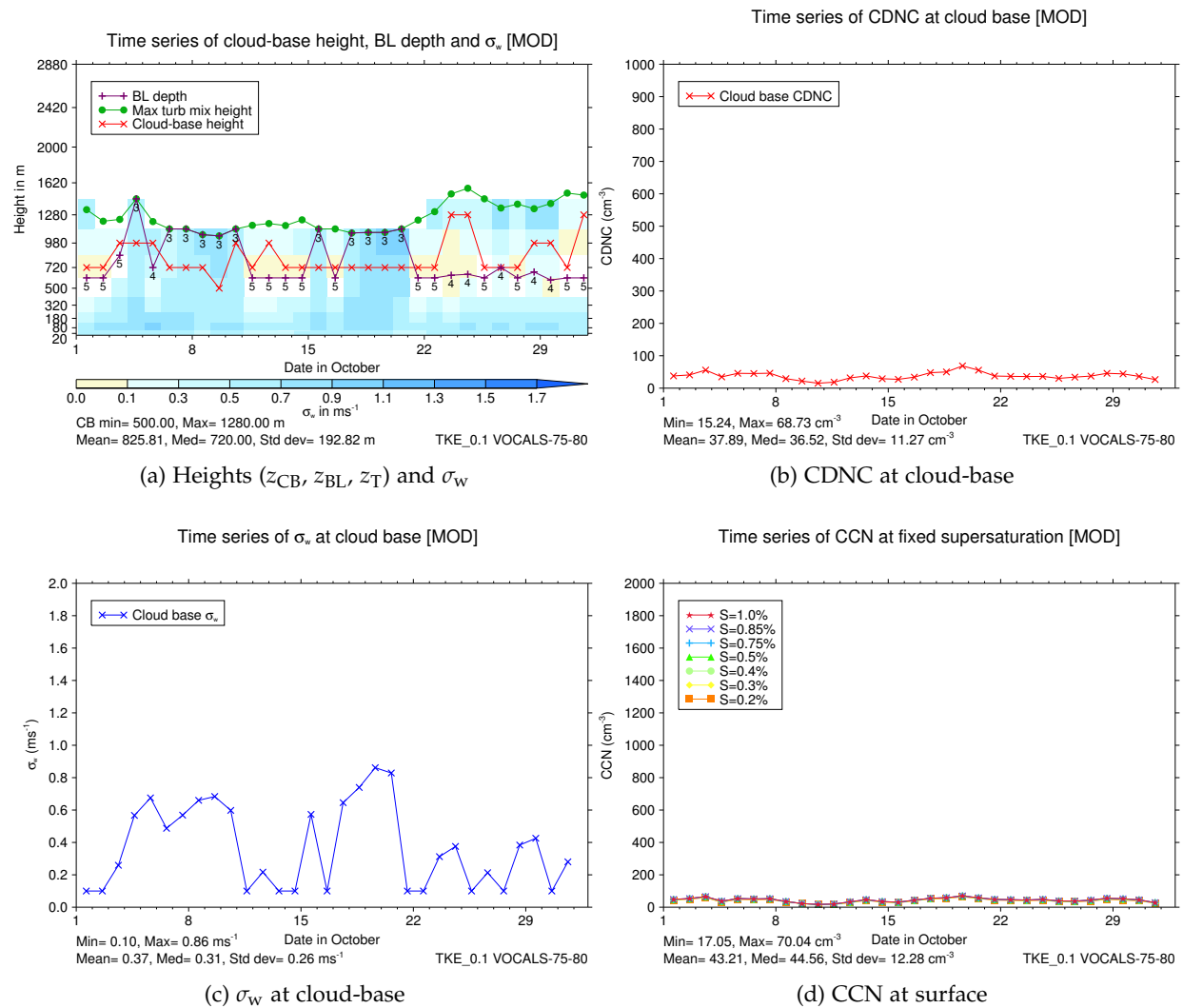


Figure 5.9: As Fig. 5.2 for model data co-located with VOCALS-75-80 i.e. the model is sampled at 77.5W, 20S.

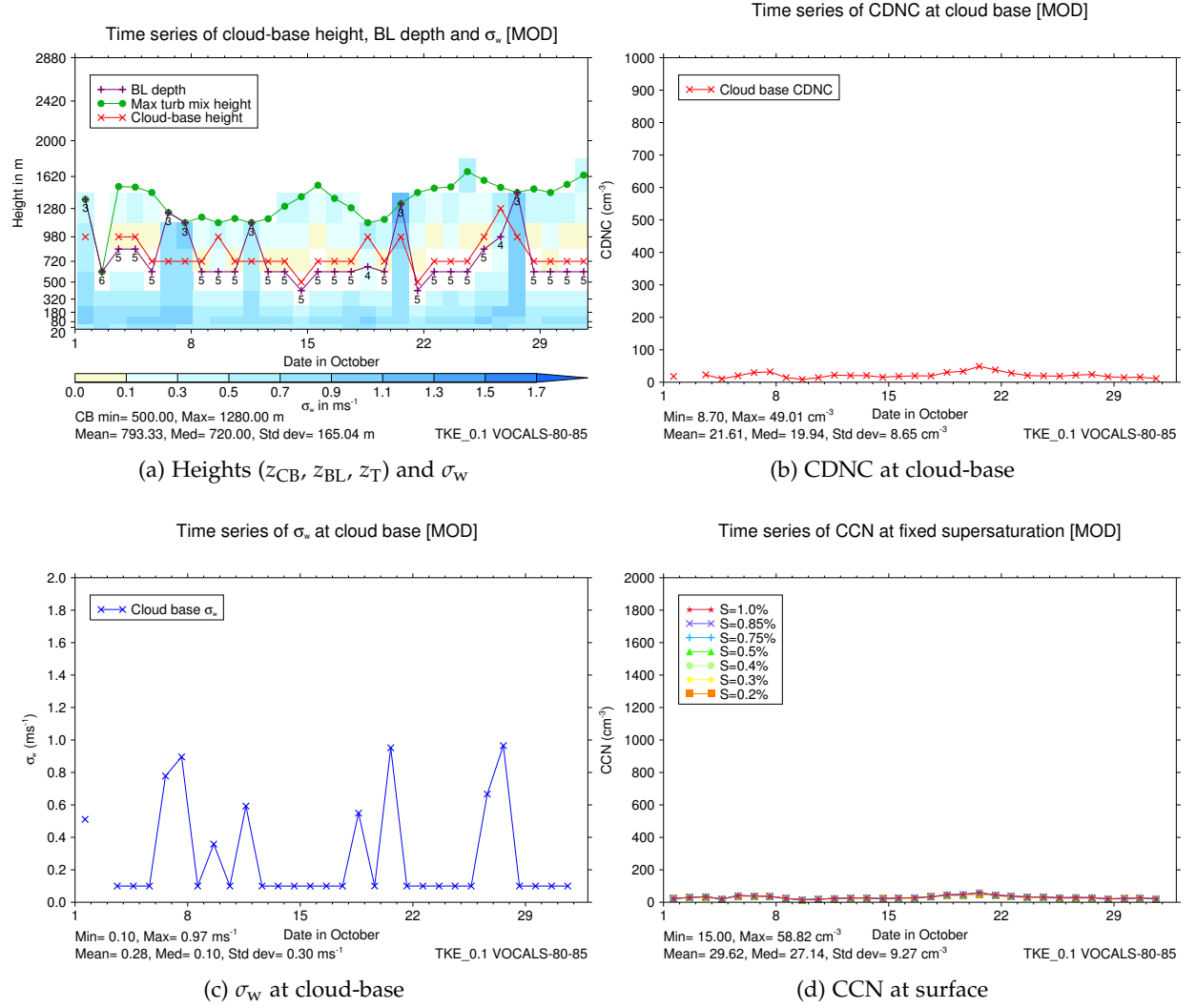


Figure 5.10: As Fig. 5.2 for model data co-located with VOCALS-80-85 i.e. the model is sampled at 82.5W, 20S.

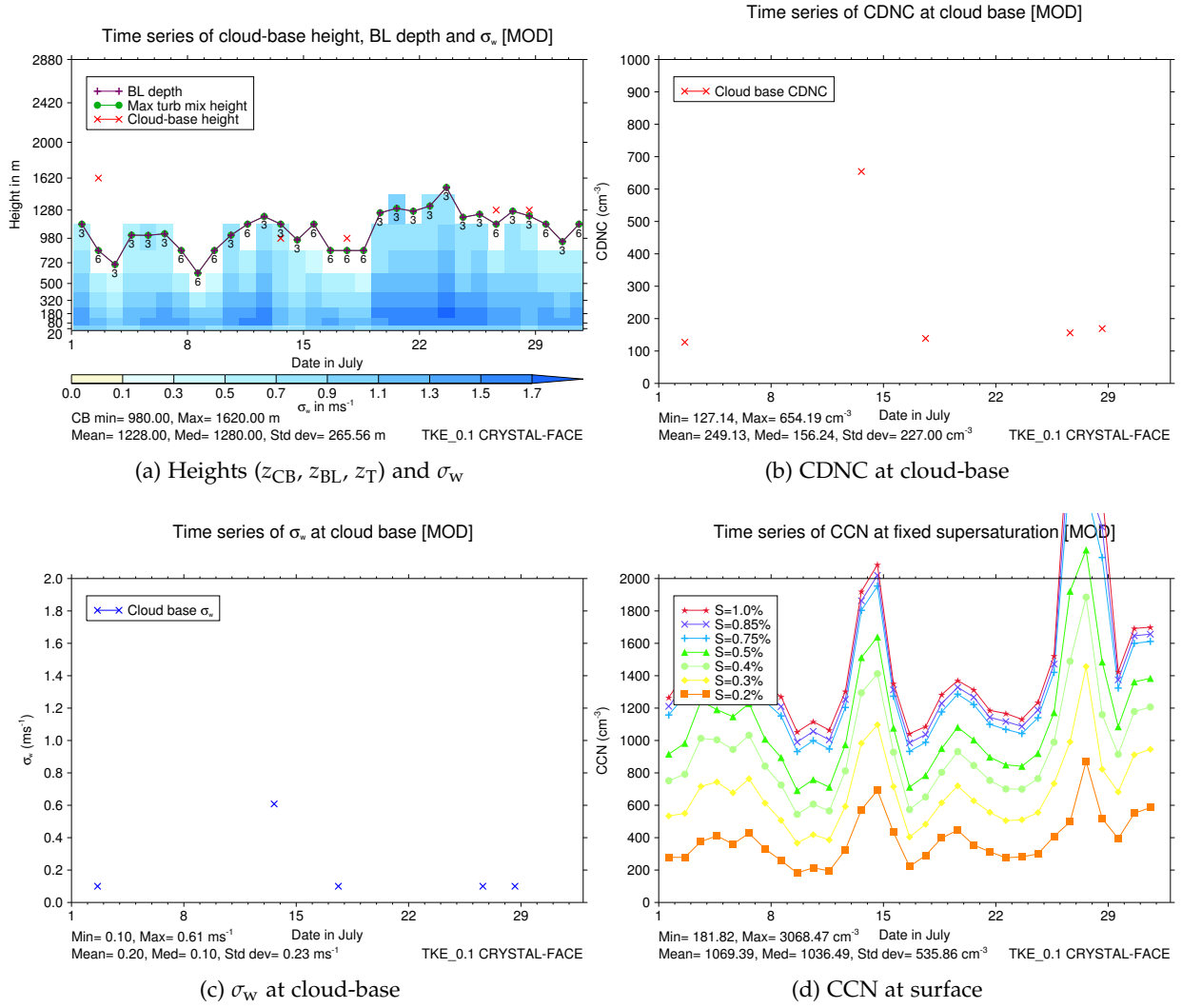


Figure 5.11: As Fig. 5.2 for model data co-located with CRYSTAL-FACE.

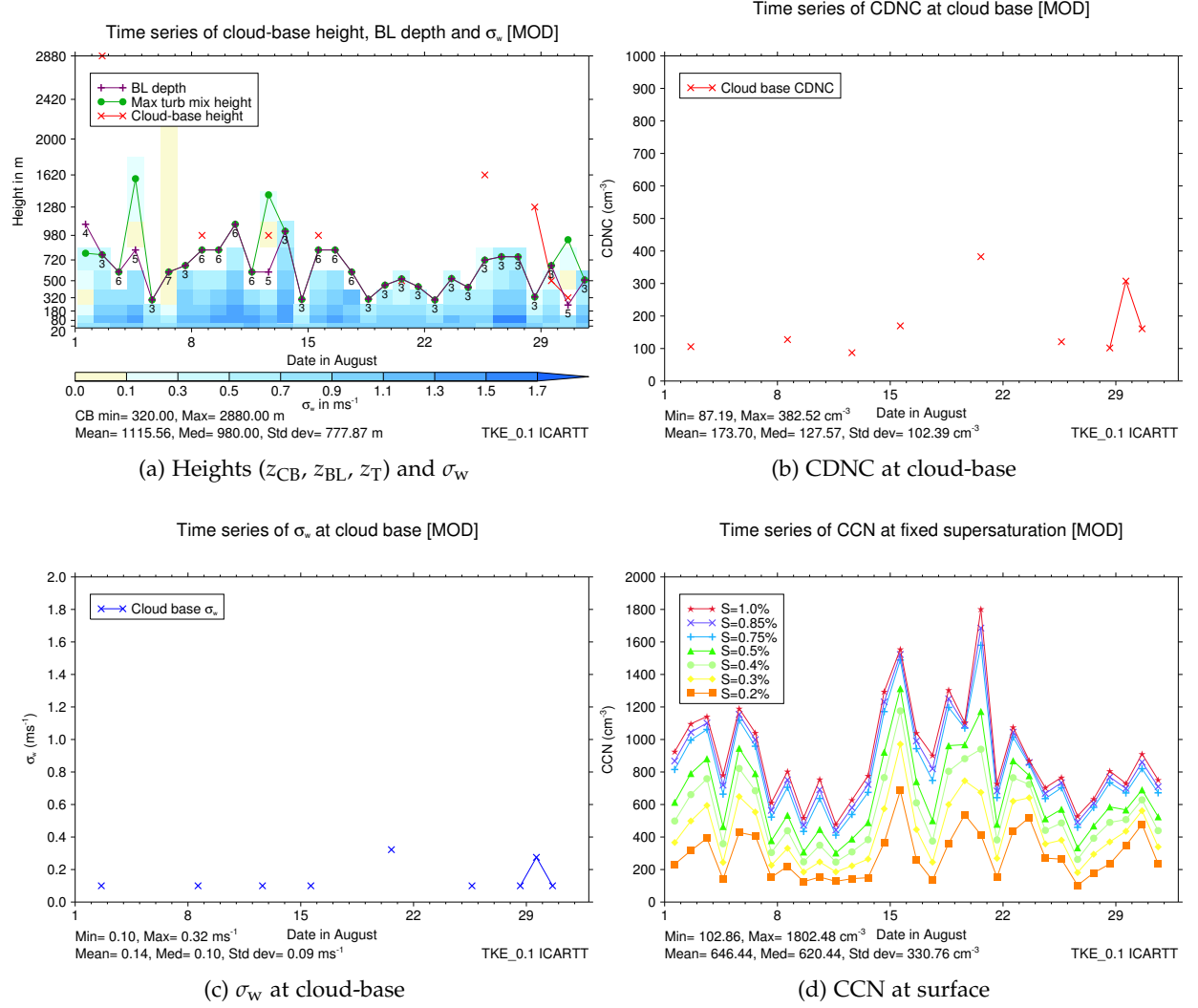


Figure 5.12: As Fig. 5.2 for model data co-located with ICARTT.

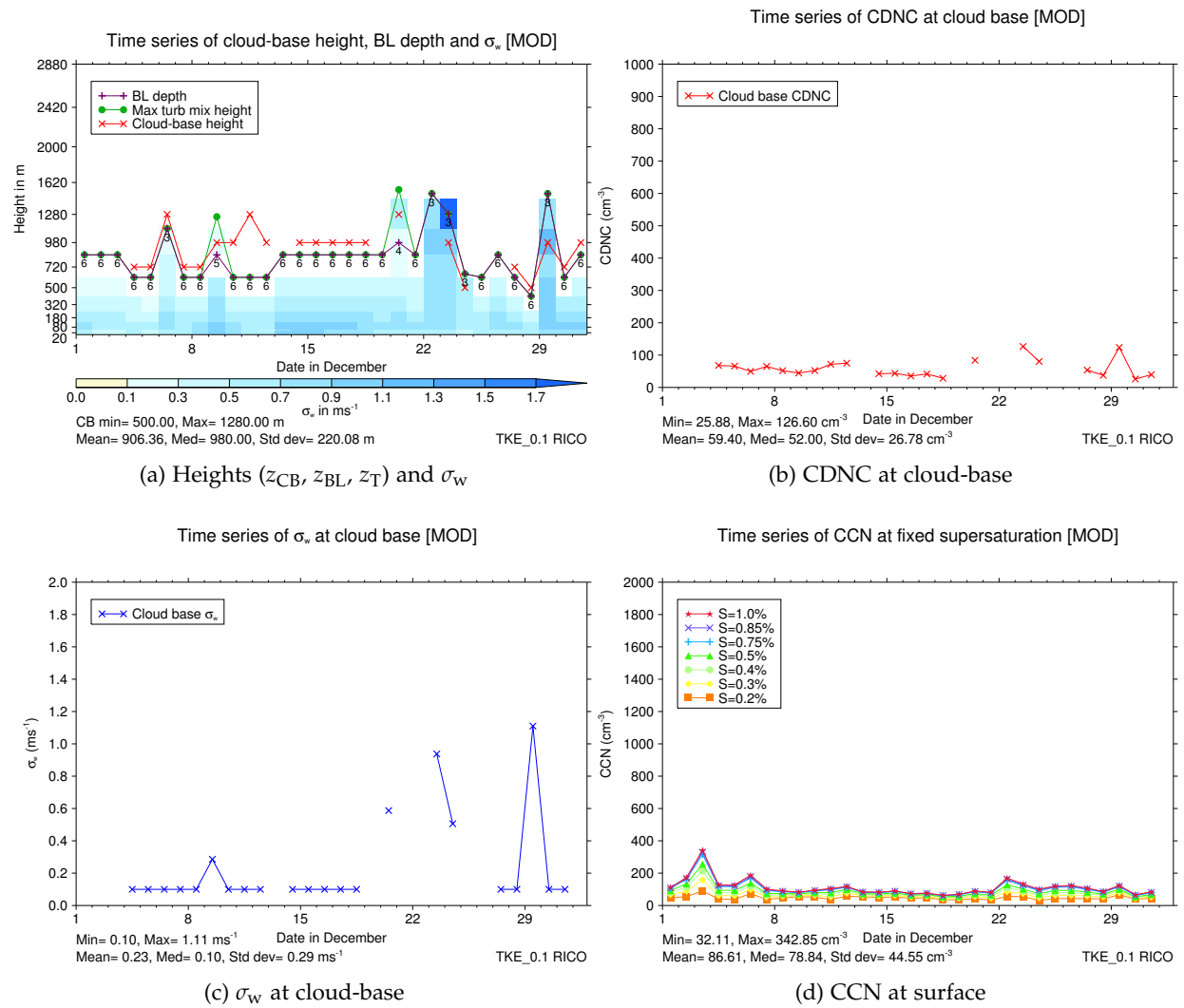


Figure 5.13: As Fig. 5.2 for model data co-located with RICO.

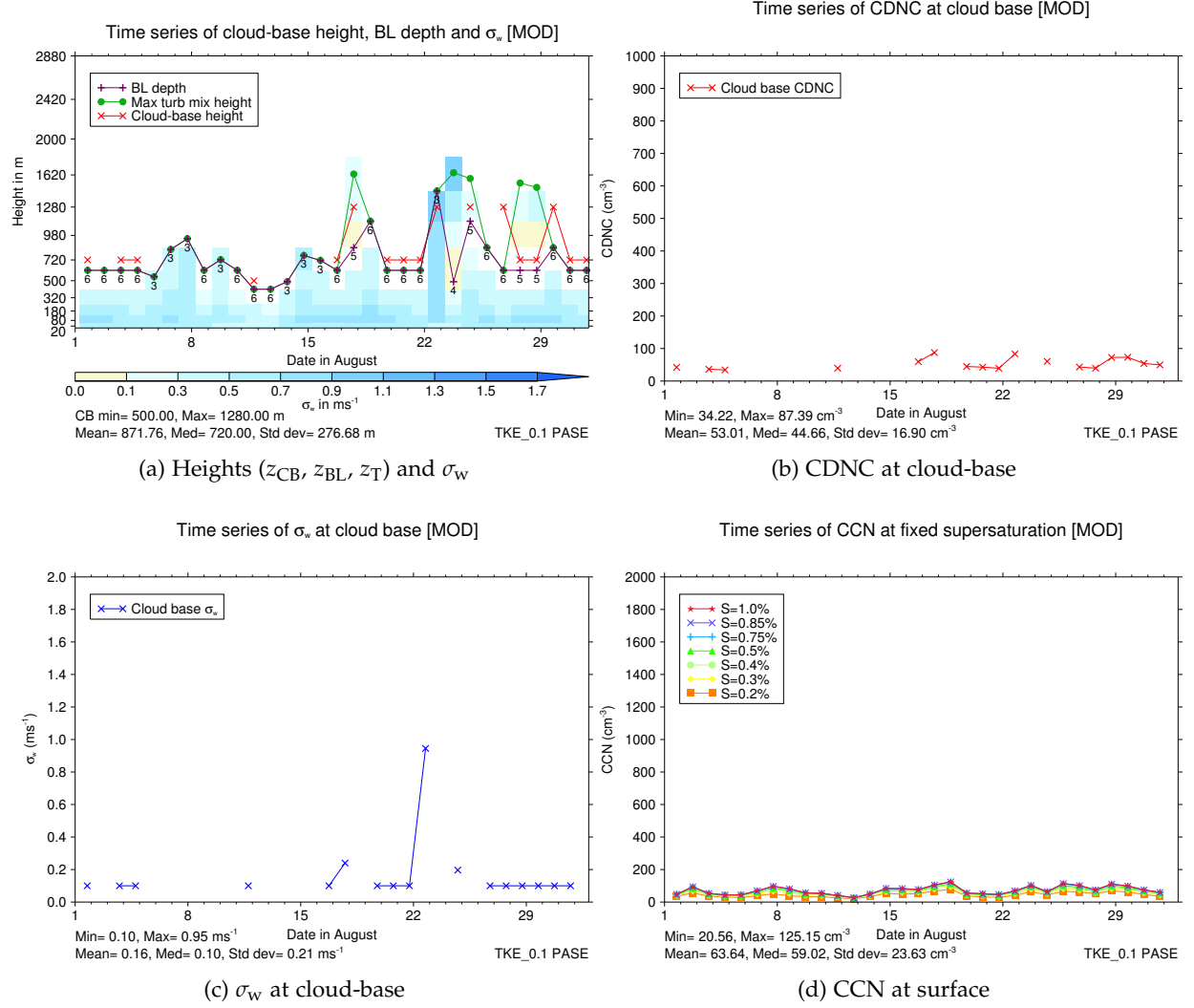


Figure 5.14: As Fig. 5.2 for model data co-located with PASE.

5.3 Comparisons between model and observations

Comparisons between model output and *in situ* flight campaign datasets require a certain number of assumptions to be made, in particular due to the limited spatial and temporal resolution of the model. As described above, the model data have been co-located as far as possible in time and space to the mean month and location of each flight campaign. Even so, conclusions from comparisons can only be meaningful to the extent that the underlying assumptions can be considered valid. In Figs. 5.15 and 5.16 the range, interquartile range and median value of CDNC and σ_w as measured throughout the course of each flight campaign are compared with those modelled instantaneously at a fixed time each day for a month in the four different model configurations listed in Table 4.2.

Published flight campaign datasets typically consist of a table of in-cloud or in-flight means of CDNC, $\bar{w}$, σ_w and aerosol number concentration (over a particular size range). In some cases measurement error estimates are also specified. Measurements are typically taken at a frequency of ~ 1 Hz so these in-cloud means incorporate thousands of individual measurements, made over flights lasting a few hours, and covering hundreds of kilometres in distance. (There is little consistency between campaigns of the length of flight legs or the selection criteria with which clouds were chosen, or the instrumentation used to make measurements.) We assume that mean CDNC over the whole flight leg is representative of conditions throughout the flight (i.e. assume low standard deviation, where this is not published), and that the measurements made along the flight track are representative of conditions throughout the cloud. For updraught velocity, we assume that published σ_w indicates the standard deviation of updraught throughout the flight, and that this is representative of all clouds within an area the size of a model grid-box.

The distribution of these in-cloud mean updraughts over the course of the flight campaign is then compared with the distribution of instantaneous model values of grid-box in-cloud σ_w and CDNC at cloud-base, obtained at a fixed time each day. In doing so, we make the assumption that the temporal sampling of one instantaneous value of a grid-box mean each day for a month from the model generates a sample which is comparable to the dataset obtained by the temporal and spatial averaging and sampling of the flight campaign.

We also assume that it is reasonable to compare flight campaign data with output from a model whose meteorological variables are nudged to ECMWF re-analysis for a different year, 2008. Finally, we note the small sample size in almost all cases of both measured and modelled data. (Sample size is noted on box-and-whisker plots.)

5.3.1 Marine stratiform clouds

Fig. 5.15 contains box-and-whisker plots showing the minimum, median, maximum and quartile values of CDNC and σ_w for each of the marine stratiform flight campaigns. For each campaign, observations are shown on the left of each group; distributions from each model configuration are plotted from left to right: TKE_0.1, sigw0.1, sigw0.4, and sigw0.7. In (b), observations of σ_w are plotted where published; distributions from TKE_0.1 are again plotted as box-and-whiskers; finally, appropriate fixed values of σ_w are plotted for experiments sigw0.1, sigw0.4, and sigw0.7, for comparison.

Model TKE_0.1 shows a good match to the observed median CDNC for NARE and CLOPAP. For NARE, the inter-quartile range (IQR) of CDNC from TKE_0.1 is very similar to the observed IQR, although median and 75th percentile of σ_w from TKE_0.1 are much higher than that observed. For CLOPAP, the 75th percentile of modelled σ_w also exceeds observations, although the median modelled σ_w is slightly lower than that observed. It is

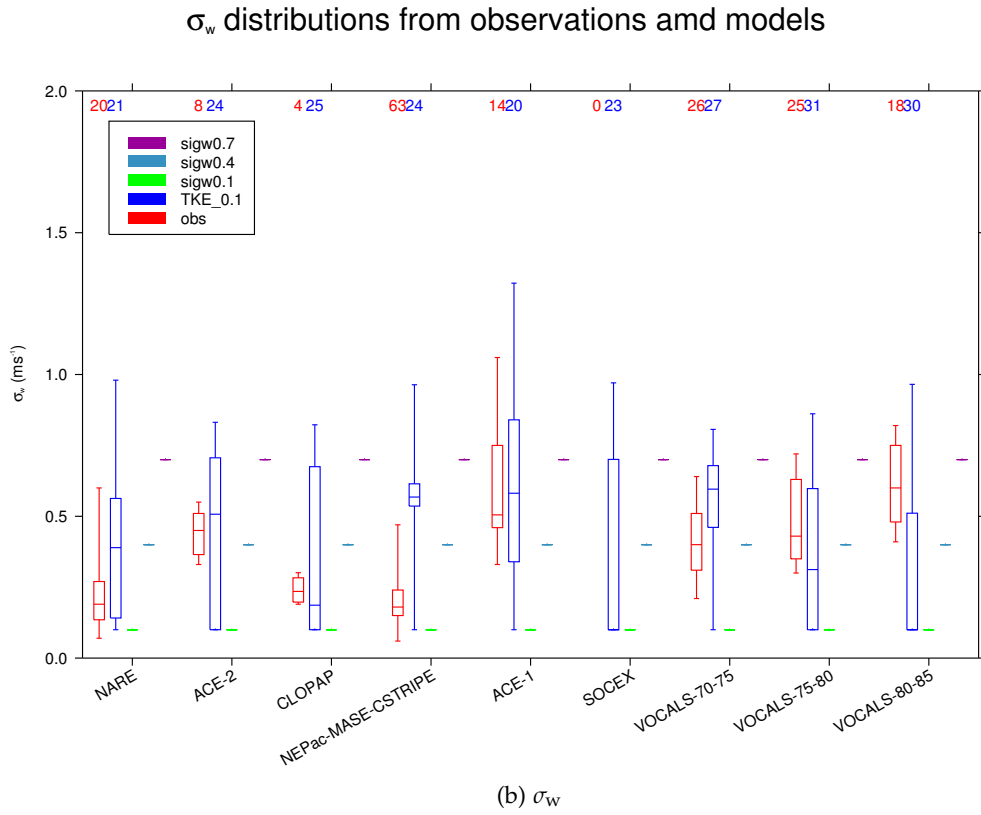
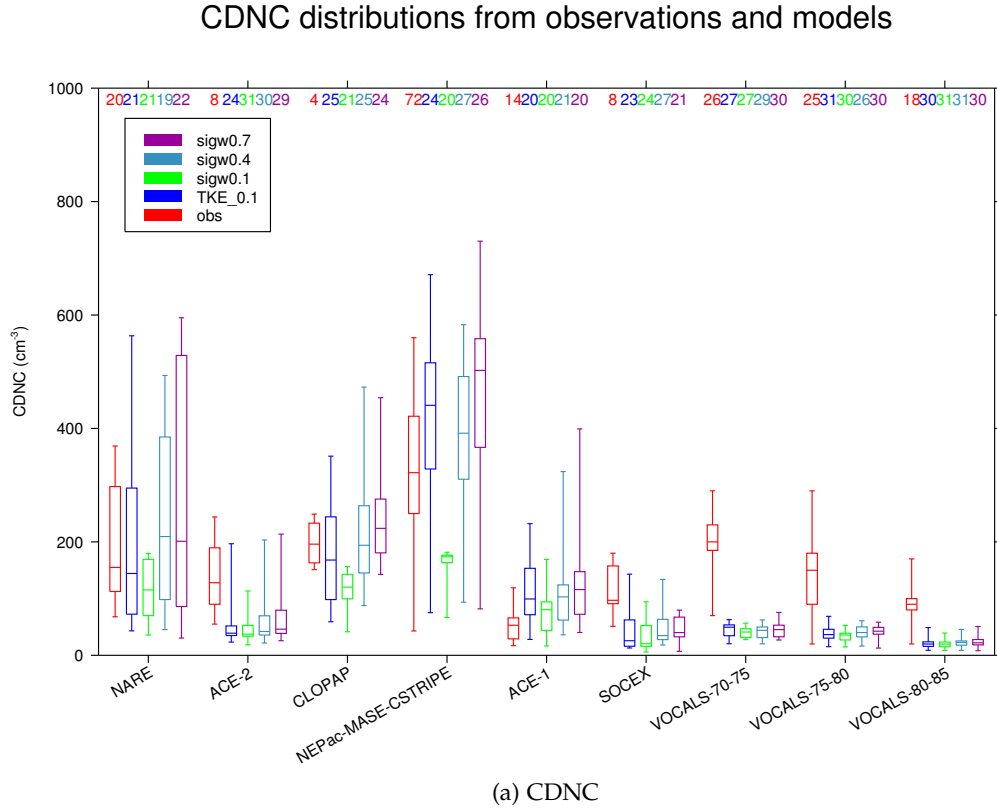


Figure 5.15: Box-and-whisker plots showing minimum, 25th percentile, median, 75th percentile and maximum values of (a) CDNC and (b) σ_w , from observations from flight campaigns (where reported) in red, and each model configuration (TKE_0.1 in blue, sigw0.1 in green; sigw0.4 in turquoise and sigw0.7 in purple) for marine stratiform flight campaigns. Note that in (b) appropriate fixed values of σ_w are plotted for experiments sigw0.1, sigw0.4, and sigw0.7, for comparison purposes.

possible for these cases that an excess of σ_w may be compensated for by a lack of CCN at these points in the model.

CDNC are slightly overestimated by TKE_0.1 compared to observations for NEPac-MASE-CSTRIPE and ACE-1. For NEPac-MASE-CSTRIPE, it is clear from (b) that modelled σ_w is too high, since the modelled median far exceeds the median of the observations, and this excess in σ_w probably leads to some of the excess CDNC. That increasing σ_w can increase CDNC in this region is also evident from the different ranges of CDNC simulated by the three model runs with fixed σ_w : sigw0.1, sigw0.4 and sigw0.7. However, modelled CCN may also be too high in this region. For ACE-1, it is likely that an excess of modelled CCN is the strongest contributing factor to the excess of CDNC, since the median CDNC from all of the model runs exceeds the median observed CDNC (even including sigw0.1, for which the prescribed σ_w is far lower than the observed median of $\sigma_w = 0.5 \text{ m s}^{-1}$).

CDNC are significantly under-estimated by TKE_0.1 for SOCEX, ACE-2, and all three VOCALS cases, compared to observations. Although the low values of σ_w —due to cloud-base falling below the decoupled stratocumulus layer shown in the model time series for some of these cases—are problematic (in particular for SOCEX, VOCALS-80-85 and VOCALS-70-75), a lack of model CCN is also a significant factor. In the VOCALS cases it is clear that low model CCN dominates the low σ_w , since even when the modelled σ_w far exceeds that observed, the number of modelled CDNC is much less than that observed. (For instance, for VOCALS-70-75, the number of CDNC modelled by sigw0.7, is much less than the number observed, even though the median observed $\sigma_w = 0.4 \text{ m s}^{-1}$.) This lack of modelled CCN has been verified with as-yet unpublished, but publicly-available profiles of CCN obtained along the 20°S transect of VOCALS-REx (not shown), in which the number concentration of CCN away from coastal sources has a value closer to 200 cm^{-3} at $S = 0.4\%$, compared to low values in the range 20 to 50 cm^{-3} as simulated by the model (J. Snider, personal communications). The lack of modelled CCN at this location in HadGEM-UKCA could simply be due to a difference between the prescribed aerosol emissions in this region (from the CMIP5 aerosol emissions inventory for the year 2000 (Lamarque et al., 2010), compared to what actually happened locally during the VOCALS-REx campaign in 2008. However, there may be other systemic reasons, and this discrepancy warrants further investigation.

5.3.2 Other clouds

Fig. 5.16 contains box-and-whisker plots of (a) CDNC and (b) σ_w for model simulations and flight campaigns through cloud regimes other than marine stratiform.

For the two tropical cases, RICO and PASE, it is clear from (b) that the median value of σ_w from model TKE_0.1 is too low. As described above, this is due to the fact that the lowest cloudy level usually occurs in the level just above z_{BL} for BL type 6 (cumulus-capped layer) and therefore σ_w is forced to take on the minimum value. However, it is necessary to increase σ_w up to at least 0.7 m s^{-1} —considerably higher than the median observed in both cases—for modelled CDNC to come close to that observed, indicating that lack of CCN is also likely to be a contributing factor to the lower median value of modelled CDNC compared to observations.

For CRYSTAL-FACE, the best match (albeit still a gross underestimate) with observed range of CDNC, is obtained by model TKE_0.1, even though TKE is not a typically good predictor for updraught velocity at the base of convective clouds, but simply because the small sample size means that one high value of σ_w and CDNC pulls up the 75th percentile to the same as that obtained by sigw0.7, and increases the maximum still higher. The

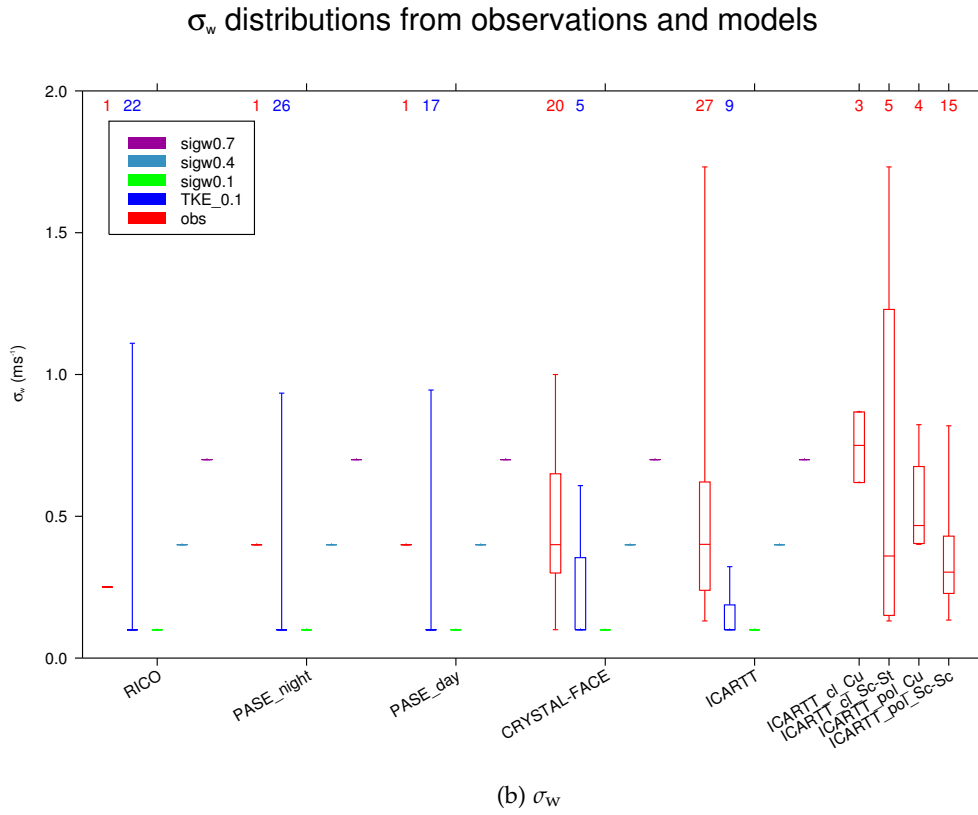
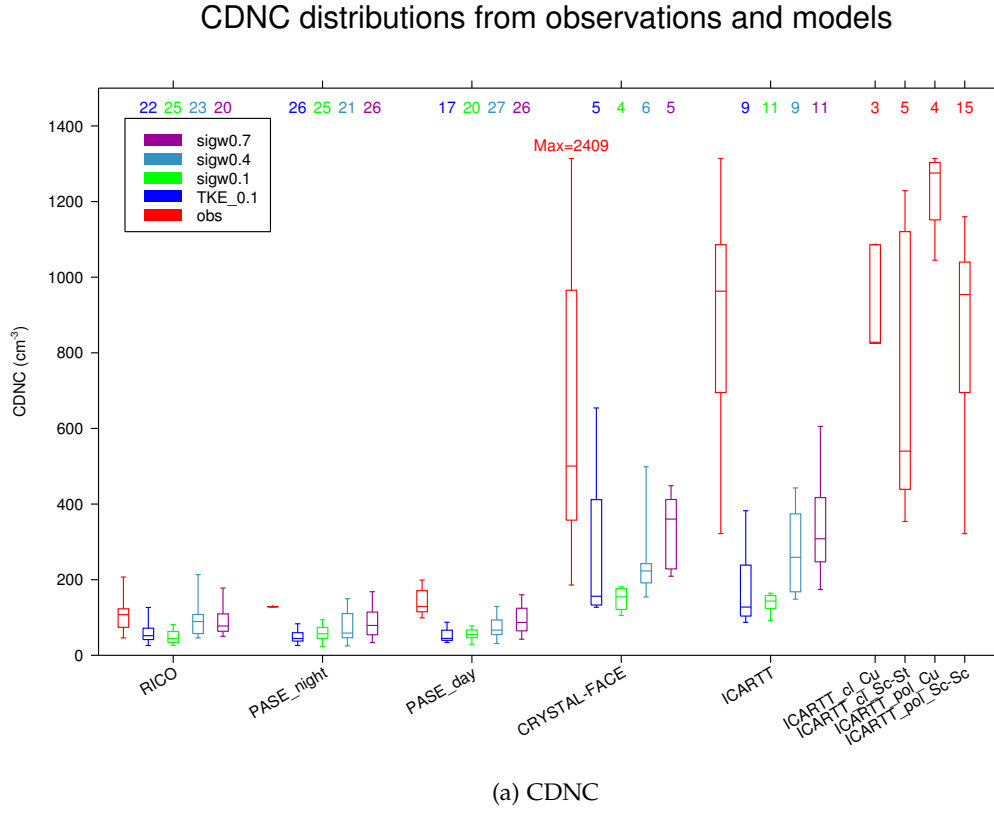


Figure 5.16: Box-and-whisker plots showing minimum, 25th percentile, median, 75th percentile and maximum values of (a) CDNC and (b) σ_w , from observations from flight campaigns (where reported) in red, and each model configuration (TKE_0.1 in blue, sigw0.1 in green; sigw0.4 in turquoise and sigw0.7 in purple) for flight campaigns through other types of cloud. Note that in (b) appropriate fixed values of σ_w are plotted for experiments sigw0.1, sigw0.4, and sigw0.7, for comparison purposes.

time series of modelled CCN for CRYSTAL-FACE [Fig. 5.11 (d)] indicates that there is no shortage of CCN in this region, but stronger updraughts are required to activate the number of droplets observed at the base of these convective clouds. Since UKCA-Activate was not designed to simulate aerosol effects on convective clouds, this is not unexpected, but it does provide evidence that a different parameterisation of vertical velocity will be needed to model aerosol activation in convective clouds.

For ICARTT, observations are split according to the cloud type recorded during the flight campaign into clean cumulus, clean stratus/stratocumulus, polluted cumulus, and polluted stratus/stratocumulus. Box-and-whisker plots for each of these subsets of the dataset are shown on the far right-hand side of Fig. 5.16. Polluted clouds of both sorts have higher median values of observed CDNC than their clean counterparts. The IQR for both clean and polluted stratiform clouds is broader and extends to lower values than for the cumulus clouds. The modelled ranges of CDNC overlap a little with the ranges for the observed stratiform clouds. In particular, median modelled CDNC for sigw0.7 is close to the median observed CDNC in clean stratiform clouds. It is not surprising that observed CDNC in the polluted stratiform clouds far exceeds that modelled, since some of these flights were conducted in close proximity to power stations, and hence higher CCN would be required for the model to be able to replicate this. As mentioned above for CRYSTAL-FACE, it is clear from (b) that far stronger updraughts are required to model the level of CDNC observed in the cumulus clouds.

5.3.3 Conclusions for UKCA-Activate

In light of this evaluation against flight campaign data, which parameterisation of vertical velocity should be recommended for the baseline configuration of UKCA-Activate? The box-and-whisker plots of Fig. 5.15 indicate that, of the limited number of marine stratiform cases shown, the sigw0.4 and TKE_0.1 results both compare reasonably well against the measurements of CDNC. As noted above, discrepancies between modelled and measured CCN at some locations may obscure the usefulness of the comparisons between modelled and measured CDNC. This makes it especially difficult to gauge which model configuration behaves best, in particular for the VOCALS, SOCEX, and ACE-2 examples. Of the remaining cases, there is no clear winner between TKE_0.1 and sigw0.4, and this is compounded by the very limited sample size.

Nevertheless, it may still be possible that the TKE_0.1 configuration performs (and could perform) better than sigw0.4 in scenarios where observed σ_w is far from the median, since the dependence of σ_w on the TKE means that it can take on a broad range of potentially more realistic values, and eliminate some of the over- and under-estimates that are bound to occur with a fixed value. Currently, the range of σ_w produced by the model is broader than that observed, as shown in Fig. 5.15 (b). However, this is partly due to the fact that nearly all cases include some frequency of occurrence of $\sigma_w^{\min}$, either due to cloud-base occurring above the maximum height of turbulent mixing, or in the gap between the two layers of turbulence in the decoupled cases (as shown in the model time-series, Figs. 5.2 to 5.8). A solution to these problems may lead to an improvement in the range of values of in-cloud σ_w used by the model, and hence also an improvement to the CDNC.

In the flight campaigns through the non-marine stratiform cases (PASE, RICO, CRYSTAL-FACE and ICARTT), none of the model configurations compare particularly well against the observations of CDNC. Sigw0.4 is the best for RICO, but for PASE, CRYSTAL-FACE and ICARTT, a lack of model CCN means that even sigw0.7 fails to simulate the observed levels of CDNC. The CDNC modelled by TKE_0.1 for the two tropical cumulus cases (RICO and

PASE) is particularly low, due to the high frequency of occurrence of $\sigma_w^{\min}$. As described above, in these cases the cloud base often seems to occur in the model level just above a type 6 boundary layer. Correcting this problem, perhaps by allowing $\sigma_w^{\min}$ to be dependent on cloud regime or boundary layer type, may enable better representation of CDNC in shallow convective clouds.

5.4 Conclusions and future work

In order to inform the best choice of σ_w for the baseline configuration of UKCA-Activate, high time-resolution model output from each vertical velocity configuration described in Chapter 4 has been co-located with and evaluated against *in situ* measurements from a dozen flight campaigns, mostly through marine stratocumulus clouds, but also in a few shallow convection cases. In the marine stratiform cases, both the sigw0.4 and TKE_0.1 configurations generally compare well against the measurements, but the sample size is not large enough to recommend one over the other. Despite this, it could perhaps be argued that the TKE_0.1 configuration is a better choice, since it is no worse than sigw0.4, and a range of σ_w is more physical than a fixed value of σ_w applied universally, as in the sigw0.4 case.

However, a potential shortcoming of the TKE_0.1 configuration is the high frequency of occurrence of $\sigma_w^{\min}$. In particular, this value is assumed to occur universally above the boundary layer. In the latter case, basing choice of $\sigma_w^{\min}$ on observations is impossible since all the flight campaigns found at the time of writing concentrate their efforts within the boundary layer. Any effect this may have on radiative forcing could be reduced in significance because many clouds above the boundary layer will be above existing lower cloudy layers.

Model time-series from TKE_0.1 indicate two main areas for the model-specific improvement of UKCA-Activate which should be addressed in future work. Firstly, it seems that the lowest cloudy level often falls between layers of turbulence for decoupled stratocumulus, such that σ_w is forced to take on the minimum value, even when there is non-zero TKE in the stratocumulus layer itself. Secondly, while none of the model configurations do particularly well in comparison to the observations through convective clouds, the model time-series indicate one way in which this could be improved, at least for shallow cumulus. In the two shallow cumulus cases studied, model cloud-base was often found to be just above the maximum height of turbulent mixing for cumulus-topped boundary layers. Allowing the value of $\sigma_w^{\min}$ to be regime-dependent may lead to improvements in the modelled CDNC. Ideally, the form of $P(w)$ should be regime-dependent, but this would require a far more comprehensive, co-ordinated, global measurement effort, in conjunction with LES studies to inform parameterisation development.

Estimation of radiative effects of anthropogenic aerosol

6.1 Radiative flux perturbations

The concept of radiative forcing was introduced in Chapter 1 as a means of quantifying and comparing the effects of isolated drivers of climate change on the Earth's radiative balance. In the definition of radiative forcing provided in the IPCC Fourth Assessment Report (Forster et al. 2007), the change in radiative flux at the tropopause is calculated instantaneously, and requires that the state of the troposphere be held constant. Thus it is possible to calculate the radiative forcing of the cloud albedo effect, but not the cloud lifetime effects, which necessarily include alterations to the state of the troposphere via changes to the hydrological cycle (e.g. through the suppression of drizzle, increasing cloud lifetime and/or cloud top height). Cloud lifetime effects cannot be diagnosed instantaneously since they are intrinsically dependent on the fast feedbacks associated with precipitation. An alternative metric is therefore required that is suitable to quantify all of the radiative effects of anthropogenic aerosols. Following the recommendations of Rotstayn and Penner (2001); Haywood et al. (2009); Lohmann et al. (2010), this chapter uses the radiative flux perturbation (RFP). Estimates of the RFP due to anthropogenic aerosol effects are derived from the difference in annual mean net radiation at the top of the atmosphere between pairs of parallel GCM simulations with present day (PD) and pre-industrial (PI) aerosol emissions.

6.1.1 Calculation of radiative flux perturbations in HadGEM-UKCA

In the simulations that follow, HadGEM-UKCA is run in atmosphere-only mode with fixed sea surface temperatures and sea-ice extent. As described in Chapter 3, the model is run in nudged mode, meaning that Newtonian relaxation—a simple form of data assimilation—is used to adjust dynamical variables in the free-running GCM towards a more realistic representation of the atmosphere at a given time, using meteorological re-analysis data. In HadGEM-UKCA, horizontal wind components (u and v) and potential temperature (θ) are nudged using the ECMWF ERA-Interim Analyses (Telford et al. 2008). All simulations in this thesis have been nudged towards time-varying fields of u , v and θ for the year 2008. The integrations in this work have been restricted to twelve-month runs with each of the

Species	PI (Tg yr ⁻¹)	PD (Tg yr ⁻¹)	PD-PI (Tg yr ⁻¹)
Black carbon fossil fuel (at surface)	0.10	3.10	3.00
Black carbon biofuel (at surface)	1.17	2.12	0.95
Organic carbon fossil fuel (at surface)	0.21	4.31	4.10
Organic carbon biofuel (at surface)	5.14	8.56	3.42
Black carbon biomass (3D emissions)	2.06	2.65	0.59
Organic carbon biomass (3D emissions)	25.52	32.99	7.47
SO ₂ emissions (at surface)	1.48	37.86	36.38
SO ₂ emissions (high level, ~1 km)	0.64	67.26	66.62

Table 6.1: Global emissions from anthropogenic sources for each species considered in this study, for pre-industrial (PI) and present-day (PD) simulations. Emissions for the years 1860 and 2000 are taken from the Coupled Model Intercomparison Project Phase 5 (CMIP5) emissions dataset (Lamarque et al., 2010).

PI and PD aerosol emissions (with a preceding, discarded, three-month spin-up period). Future work could include longer simulations of five or perhaps ten years.

Aerosol emissions for the years 1860 and 2000 are taken from the Coupled Model Intercomparison Project Phase 5 (CMIP5) emissions dataset (Lamarque et al., 2010), provided to support modelling activities for the IPCC fifth assessment report (AR5). For biomass burning emissions, grass-fire emissions are added to the surface layer, and emissions from forest fires are assumed to be equally distributed between model levels two and twelve (approximately from 50 m up to 3 km). Dimethylsulfide (DMS) emissions are estimated from the interactive scheme in the Unified Model, and used to drive UKCA. A summary of the total global anthropogenic emissions relevant to aerosols in the pre-industrial and present day runs is given in Table 6.1.

Model configurations

RFPs are calculated for each of the five vertical velocity configurations used in Chapter 4 (summarised in Table 4.2). Four of the configurations use a Gaussian pdf of vertical velocity, centred on the large-scale grid-box mean vertical velocity. Of these, the standard deviation of the distribution is fixed for three of the configurations (i.e. $\sigma_w = 0.1 \text{ m s}^{-1}$ for **sigw0.1**, and so on for **sigw0.4** and **sigw0.7**). For the fourth, this standard deviation is approximated by a function of the turbulent kinetic energy in each grid-box, $\sigma_w = \sqrt{2/3 \text{TKE}}$, where this is greater than a minimum value of $\sigma_w^{\min} = 0.1 \text{ m s}^{-1}$. Above the planetary boundary layer, TKE is not defined within the model, and the minimum value is used. This configuration is referred to as **TKE_0.1**. The final configuration does not represent sub-grid-scale variability with a pdf or by any means, but simply uses a single fixed value of vertical velocity at all grid-points of $w = 0.32 \text{ m s}^{-1}$, and is referred to as **w_0.32**.

The range of RFPs from these configurations provides some indication of the uncertainty in estimates of the indirect aerosol effects due to inaccuracies in the representation of sub-grid-scale vertical velocity.

In addition to the five configurations of vertical velocity introduced in Chapter 4, **JonesCDNC** refers to results from a pair of model runs in which the Jones et al. (1994) empirical relation is used to estimate CDNC from the aerosol number concentrations predicted by UKCA, in place of the Abdul-Razzak and Ghan (2000) aerosol activation scheme. Within UKCA, the number of soluble aerosols larger than a prescribed threshold

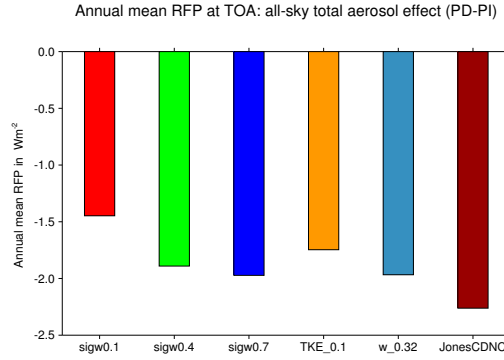


Figure 6.1: Annual mean RFPs due to total anthropogenic aerosol effects for model configurations: sigw0.1 (red), sigw0.4 (green), sigw0.7 (blue), TKE_0.1 (orange), w_0.32 (light blue), and JonesCDNC (dark red).

dry radius of 35 nm is calculated by integrating the number-size distribution of each of the soluble log-normal aerosol modes and summing over all modes to give N_{r35} . The cloud droplet number concentration in each grid-box is then estimated following Jones et al. (1994),

$$N_d = \max \left[3.75 \times 10^8 \left\{ 1 - \exp \left(-2.5 \times 10^{-9} N_{r35} \right) \right\}, N_{\min} \right] \text{ m}^{-3}, \quad (6.1)$$

where $N_{\min} = 5 \times 10^6 \text{ m}^{-3}$ is the minimum imposed cloud droplet number concentration as used by all of the configurations of UKCA-Activate.

6.2 Results

All of the configurations of HadGEM-UKCA used in this thesis predict an overall negative RFP due to anthropogenic aerosols, with estimates of the total global mean RFP ranging from -1.4 to -2.3 W m^{-2} as shown in Fig. 6.1. This range of values is notably high as compared to the observed positive forcing due to greenhouse gases and ozone of $+2.9 \pm 0.3 \text{ W m}^{-2}$ (Forster et al., 2007). Inverse estimates of aerosol forcings reported in the Fourth Assessment Report of the IPCC suggest that the net aerosol forcing over the 20th century should be negative with likely ranges between -1.7 and -0.1 W m^{-2} , to be consistent with global warming (Hegerl et al., 2007).

There are several possible reasons why the range of values obtained with HadGEM-UKCA shown in Fig. 6.1 might exceed the inverse estimates, which could be tested by further model simulations and analysis.

A previous study by Hoose et al. (2009) looked at the effect that imposing a minimum value of CDNC ($N_{\min}$) had on overall forcing estimates. Hoose et al. found that reducing this lower bound increased the magnitude of the indirect aerosol effect since perturbations from a lower value of $N_{\min}$ cause a greater relative change in CDNC and hence stronger forcing than when a high minimum value is imposed. HadGEM-UKCA imposes a minimum value of 5 cm^{-3} , which is lower than other models and this may lead to indirect effect estimates which are higher than other models. However, the conclusion of the Hoose et al. study is that it is unphysical to depend upon a high prescribed minimum value of CDNC to force models to generate indirect effects which are within the range predicted by

inverse calculations, which is exactly the reason why such a low value of $N_{\min}$ is chosen for HadGEM.

One of the limitations of the configuration of HadGEM-UKCA used for all the simulations in this thesis is the lack of a two-moment cloud scheme with prognostic cloud droplet number. The current diagnostic scheme estimates CDNC from sources of droplets only, and fails to account for evaporation of droplets or any other sink processes. It will be interesting to see how estimates of the aerosol effects change when prognostic cloud droplet number is available.

As discussed in Chapter 3, present day CCN concentrations are biased low in the model, compared to surface observations, particularly in the stratocumulus regions, and modelled aerosol optical depth is slightly lower than that observed by satellites in most regions (see Fig. 3.3). Low background aerosol concentrations from UKCA could be one of the major causes of the strong indirect aerosol effects found in the model, since the relative increase in present-day CDNC is greater when starting from a lower pre-industrial background value, which in turn leads to a stronger indirect effect.

Several changes to UKCA planned for the near future may also alter the strength of the indirect effects, such as the inclusion of dust, nitrates and further organic aerosol species and again, it will be interesting to see how these changes affect the overall estimates of the radiative effects of aerosols.

The majority of the range of RFPs predicted by the different vertical velocity configurations explored in this study (-1.4 to -2.0 W m^{-2}) is due to varying the standard deviation of the pdf of updraught velocity, demonstrating how great an impact this one parameter can have on the overall RFP.

It is important to state that in terms of their use as indicators of the effects of aerosol on climate, it is only valid to consider the *global mean* value of RFP, since it is not possible to assess the spatial distribution of the ultimate climate impacts of perturbations to radiative flux (and the hydrological cycle) simply from the regions where this RFP appears strongest. However, maps of RFP are included in Fig. 6.2, and Fig. 6.3 displays difference plots of RFPs between selected combinations of model configurations, to highlight where differences manifest themselves in this climatically important quantity.

The most striking features of the maps of RFP in Fig. 6.2 are the regions of strong negative RFP, covering much of the ocean in the northern hemisphere. The magnitude of the effect increases with increasing σ_w . Over land regions, the RFP is also predominantly negative, but less so than over ocean, other than in eastern Europe and sweeping out over China into the Pacific outflow region. In these areas the negative effect also intensifies significantly with increasing σ_w [evident in the difference plot of $\text{RFP}(\text{sigw}0.7) - \text{RFP}(\text{sigw}0.1)$ in Fig. 6.3], and is found to be similarly strong in the non-fixed- σ_w runs.

In the southern hemisphere, all model configurations show strong localised negative effects off the coast of Chile in the south east Pacific ocean and also along the coast of southern Africa. However, other than in these regions, much of the southern hemisphere (and tropical) oceans are covered by a weak and noisy positive RFP, which raises some questions. What causes the sign difference between the two hemispheres? In particular, what causes the region of elevated positive RFP in the southeastern Pacific? A comparison between the model configurations indicates that the intensity of the effect in this region increases with increasing σ_w [again, evident in the difference plot of $\text{RFP}(\text{sigw}0.7) - \text{RFP}(\text{sigw}0.1)$ in Fig. 6.3] and is strongest in configuration w_0.32. This will be investigated further in Sec. 6.3.

In the global mean, the RFP for TKE_0.1 is lower than that for sigw0.4. This relates back to one of the findings of Chapter 4, that although there are several regions where σ_w

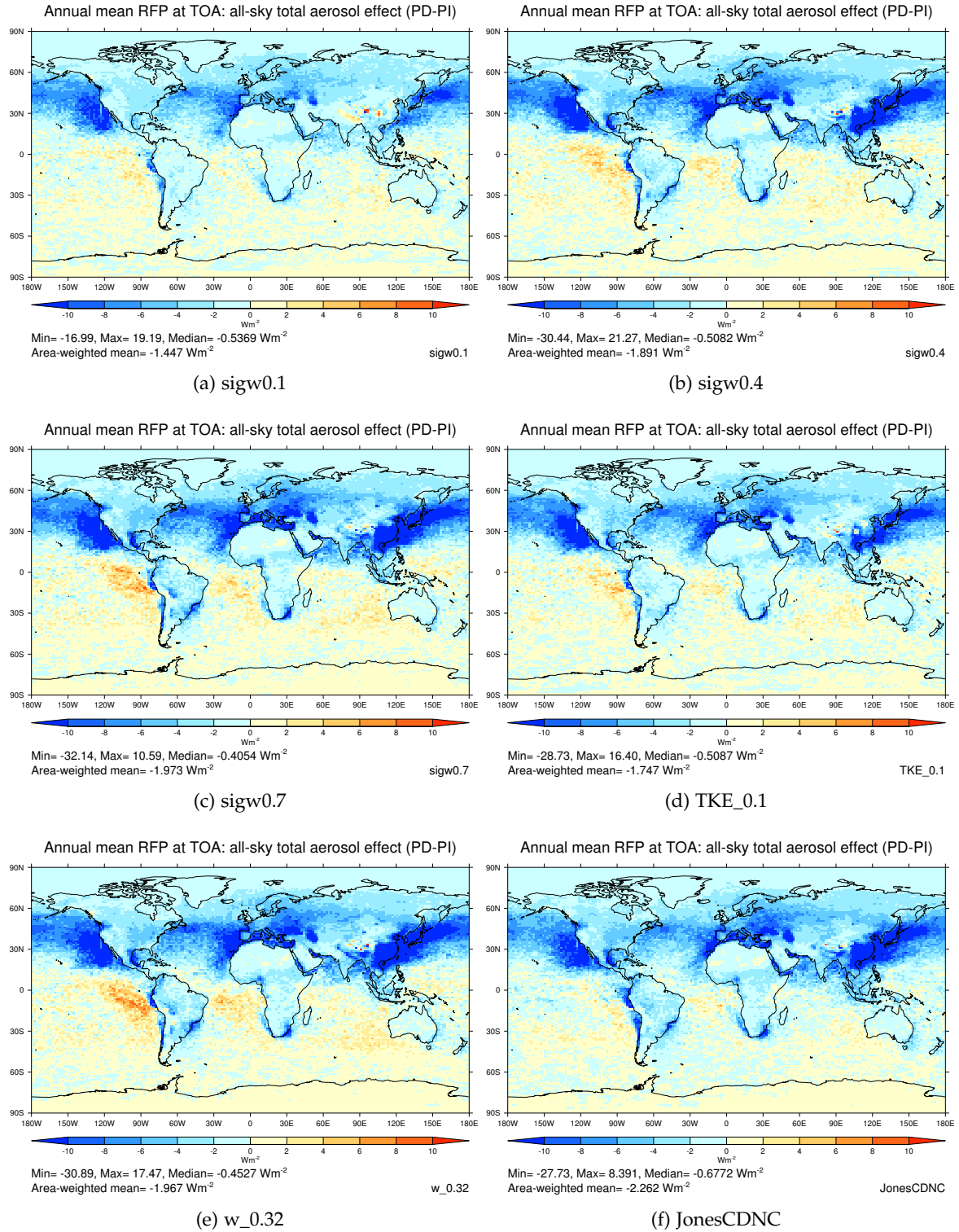


Figure 6.2: Annual mean radiative flux perturbations due to total (direct and indirect) anthropogenic aerosol effects for runs (a) sigw0.1, (b) sigw0.4, (c) sigw0.7, (d) TKE_0.1, (e) w_0.32, and finally (f) JonesCDNC (a pair of runs using the Jones et al. (1994) empirical relation between UKCA aerosol and CDNC, rather than the Abdul-Razzak and Ghan activation scheme).

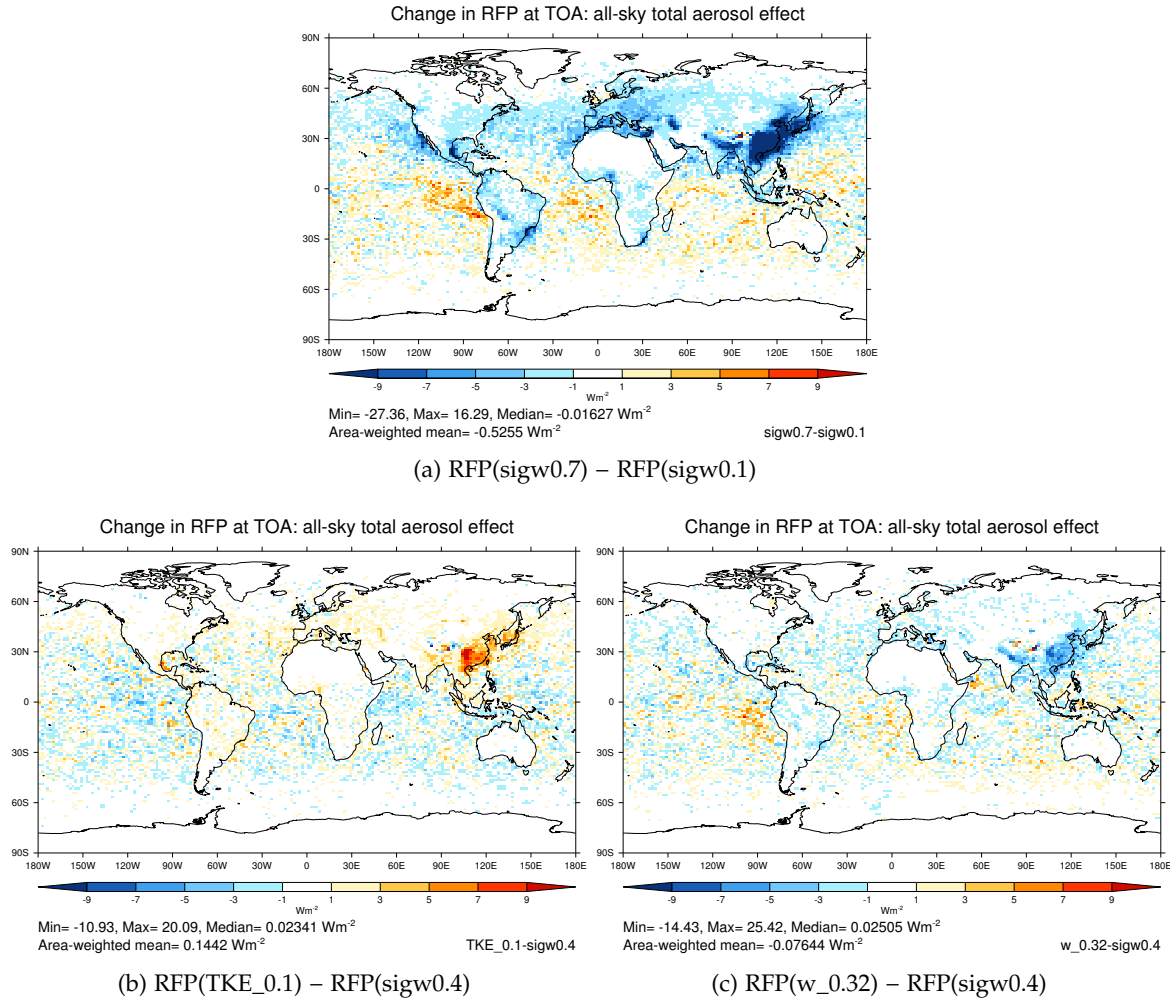


Figure 6.3: Difference in annual mean radiative flux perturbations due to total (direct and indirect) anthropogenic aerosol effects between (a) sigw0.7 – sigw0.1, (b) TKE_0.1 – sigw0.4 and (c) w_0.32 – sigw0.4.

derived from TKE exceeds 0.4 m s^{-1} (as shown for runs of TKE_0.1 compared to sigw0.4 with present-day aerosol in Fig. 4.12), the high frequency of occurrence of the minimum value of $\sigma_{w(\min)}$ in TKE_0.1 brings down the average value of σ_w . Both positive and negative features of the map of RFP for TKE_0.1 are reduced in intensity, compared to sigw0.4, for much the same reasons as they are for sigw0.1 compared to sigw0.4 (albeit to a lesser extent). Although allowing σ_w to depend on TKE produces some regions in which $\sigma_w > 0.4 \text{ m s}^{-1}$, this does not lead to noticeable increases in the CDNC compared to that obtained with sigw0.4 (illustrated in Fig. 4.13 compared to Fig. 4.12 for present-day aerosol), because in these regions CDNC tends to be limited by the number of available CCN rather than updraught. This is particularly true in the marine stratocumulus regions.

Configuration w_0.32 uses a single fixed value of vertical velocity derived from the mean value of the positive branch of the sigw0.4 pdf, $w_+ \approx 0.8\sigma_w = 0.32 \text{ m s}^{-1}$ [see Eq. (4.12)]. This produces higher CDNC and a slightly stronger RFP than sigw0.4, which is consistent with the prediction of Morales and Nenes (2010), that using w_+ would overestimate CDNC compared to using a full pdf. The reason for this can be demonstrated by referring back

to the definition of characteristic updraught in Chapter 4, i.e. that w^* is the value of w for which $N_a(w^*) = \overline{N_a}$. Morales and Nenes assume a linear relationship between the characteristic updraught and the standard deviation of updraught, $w^* = \lambda\sigma_w$, so choosing $w_+ \approx 0.8\sigma_w$ is the same as assuming a fixed value of $\lambda = 0.8$ everywhere. Again referring back to Chapter 4, Fig. 4.2 shows that for HadGEM-UKCA in configuration sigw0.4, the value of λ exhibits strong spatial variation, with a maximum value of 0.72 and a global mean value of 0.4. Thus using the mean value of the positive branch of the pdf, $w_+ \approx 0.8\sigma_w$ leads to a slight overprediction of CDNC in the PI simulations of w_0.32 compared to sigw0.4, and a greater absolute overprediction in the PD simulation (due to the greater number of CCN in present-day conditions) and hence an increased RFP.

The map of RFP in configuration JonesCDNC shows stronger negative effects in the northern hemisphere compared to sigw0.4, but weaker positive regions in the southern hemisphere, such that the effect overall is a stronger negative RFP. The differences in the intensity, patterns and balance of effects in sigw0.4 compared to JonesCDNC demonstrate the effects of the change from diagnosing CDNC simply from the number of soluble aerosols larger than a certain threshold, to using an aerosol activation scheme which can also take account of aerosol composition and competition between droplets for limited supersaturation. JonesCDNC tends to overpredict the CDNC compared to sigw0.4 in regions of high soluble aerosol concentration, hence the increase in the intensity of negative RFP in the northern hemisphere. In the southern hemisphere, the threshold radius above which JonesCDNC aerosols activate may to some extent buffer the intensity of the positive effect seen in the other configurations (explained below).

Next we consider in more detail the differences introduced by the different configurations of vertical velocity. For configurations with fixed σ_w , the magnitude of both negative and positive changes increases with increasing σ_w . As seen in Sec. 4.3, increasing σ_w tends to increase CDNC, since the increased maximum supersaturations provided by the higher updraught velocities possible with a wider pdf enable more of the smaller aerosols to activate. The increase in CDNC is particularly strong in regions of high CCN concentrations, i.e. ΔCDNC is large and positive. The same reasoning applies to intercomparisons of the RFPs between the fixed σ_w runs. The global mean CDNC in the PI run of sigw0.4 is greater than it is in the PI run of sigw0.1 but, moreover, the higher CCN concentrations in the PD runs leads to much more of an increase in CDNC for the sigw0.4 run than for sigw0.1 [i.e. $\Delta\text{CDNC}(\text{sigw0.4}) > \Delta\text{CDNC}(\text{sigw0.1})$], because the higher updraughts (and hence higher supersaturations) mean that more of the greater number of CCN in the PD run can activate, and hence the indirect radiative effects are stronger.

Fig. 6.4 shows the absolute values of CDNC generated by the reference model with (a) pre-industrial and (b) present-day aerosol emissions. The effect of increasing σ_w on ΔCDNC described above is illustrated by comparing the difference plots shown here for sigw0.4 with those in Fig. 6.5 for sigw0.1, sigw0.7 and TKE_0.1. Relative differences are shown as well as absolute differences, since it is the relative change in CDNC, combined with the cloud fraction over time, which determines the magnitude of the indirect radiative effects (as can be seen by comparing the spatial extent of, say, the negative regions of RFP in Fig. 6.2 with the corresponding relative changes in CDNC). However, again, as shown in Figs. 6.4 and 6.5, this response of ΔCDNC to increasing σ_w is sub-linear, and the increase in magnitude of RFP from sigw0.4 to sigw0.7 (-1.89 to -1.97 W m^{-2}) is less than the increase from sigw0.1 to sigw0.4 (-1.44 to -1.89 W m^{-2}).

Perturbations to the cloud liquid water path between runs with pre-industrial and present-day aerosol emissions (ΔLWP) are shown in Fig. 6.6 for sigw0.1, sigw0.4, sigw0.7 and TKE_0.1. In all four model configurations, the spatial distribution of the regions of

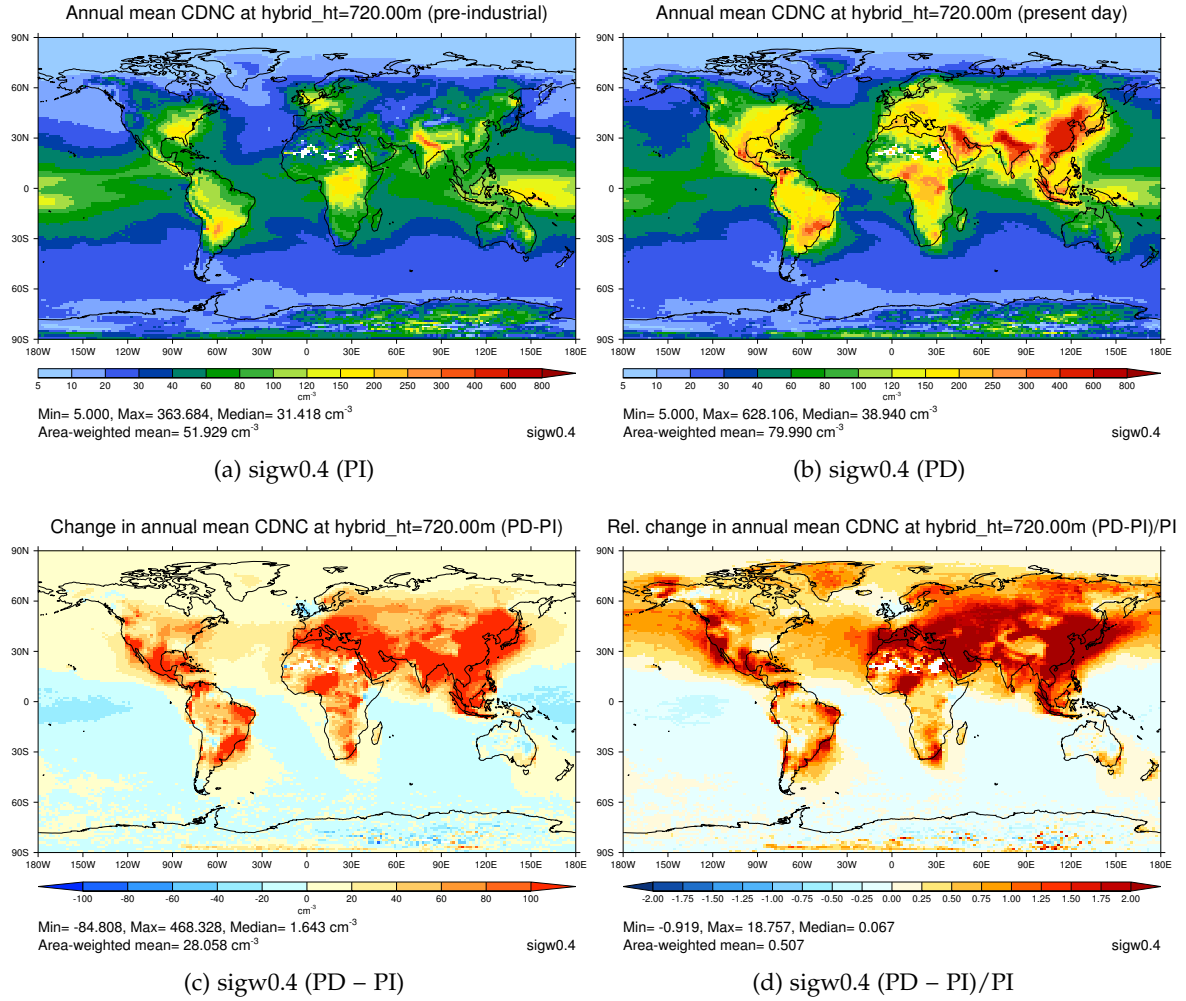


Figure 6.4: Annual mean CDNC at hybrid height = 720 m for reference model sigw0.4, run with (a) pre-industrial and (b) present day aerosol emissions; (c) shows absolute differences (PD – PI); (d) shows relative differences (PD – PI)/PI.

positive ΔLWP , corresponds well with regions of negative RFP in Fig. 6.2, particularly over ocean in the northern hemisphere. This is commensurate with the presence of additional CCN leading to an increase in the CDNC, which reduces the precipitation efficiency of a cloud and increases the cloud liquid water content. This in turn contributes to the overall negative radiative effect shown in Fig. 6.2. (By the same mechanism, the opposite of this effect can be seen over ocean in the southern hemisphere, where negative values of ΔLWP correspond to negative values of ΔCDNC shown in Fig. 6.5 and hence positive values of RFP in Fig. 6.2.)

Fig. 6.6 shows that ΔLWP increases in magnitude with increasing σ_w . Again, this is associated with the increasing magnitude of ΔCDNC with increasing σ_w shown in Fig. 6.5 and serves as a reminder that it is the primary increase in CDNC which is the basis for the secondary effects on LWP due to impacts on the precipitation efficiency, simulated in the model by construction of the autoconversion rate.

Difference plots of CDNC and LWP for TKE_0.1 have been included alongside those for the fixed σ_w models in Figs. 6.5 and show that the annual mean effects on cloud properties of

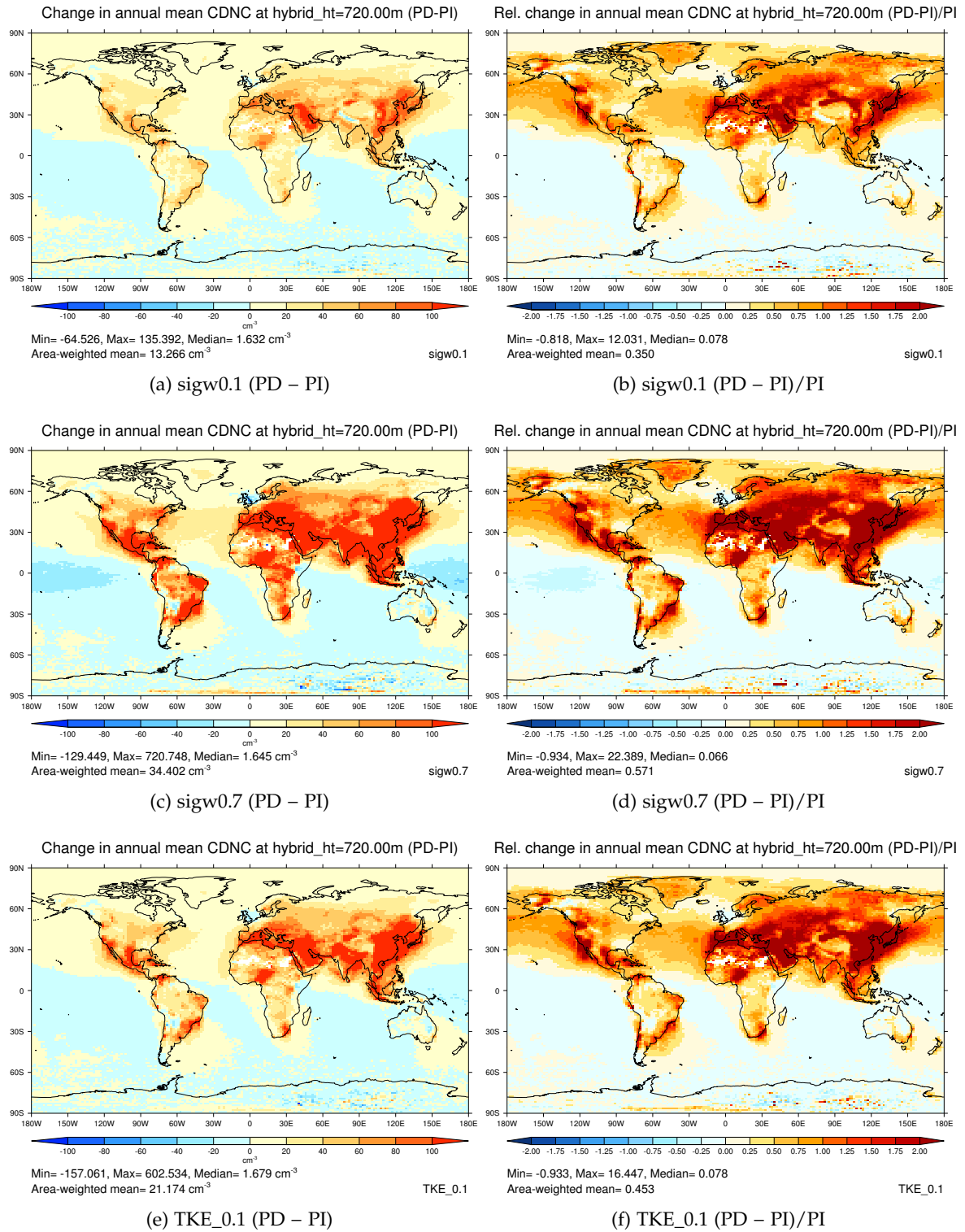


Figure 6.5: Similar to Fig. 6.4 (d) and (e): left-hand column shows absolute differences (PD - PI), and right-hand column shows relative differences (PD - PI)/PI, in annual mean CDNC at hybrid height = 720 m for model runs (a-b) sigw0.1, (c-d) sigw0.7, and (e-f) TKE_0.1.

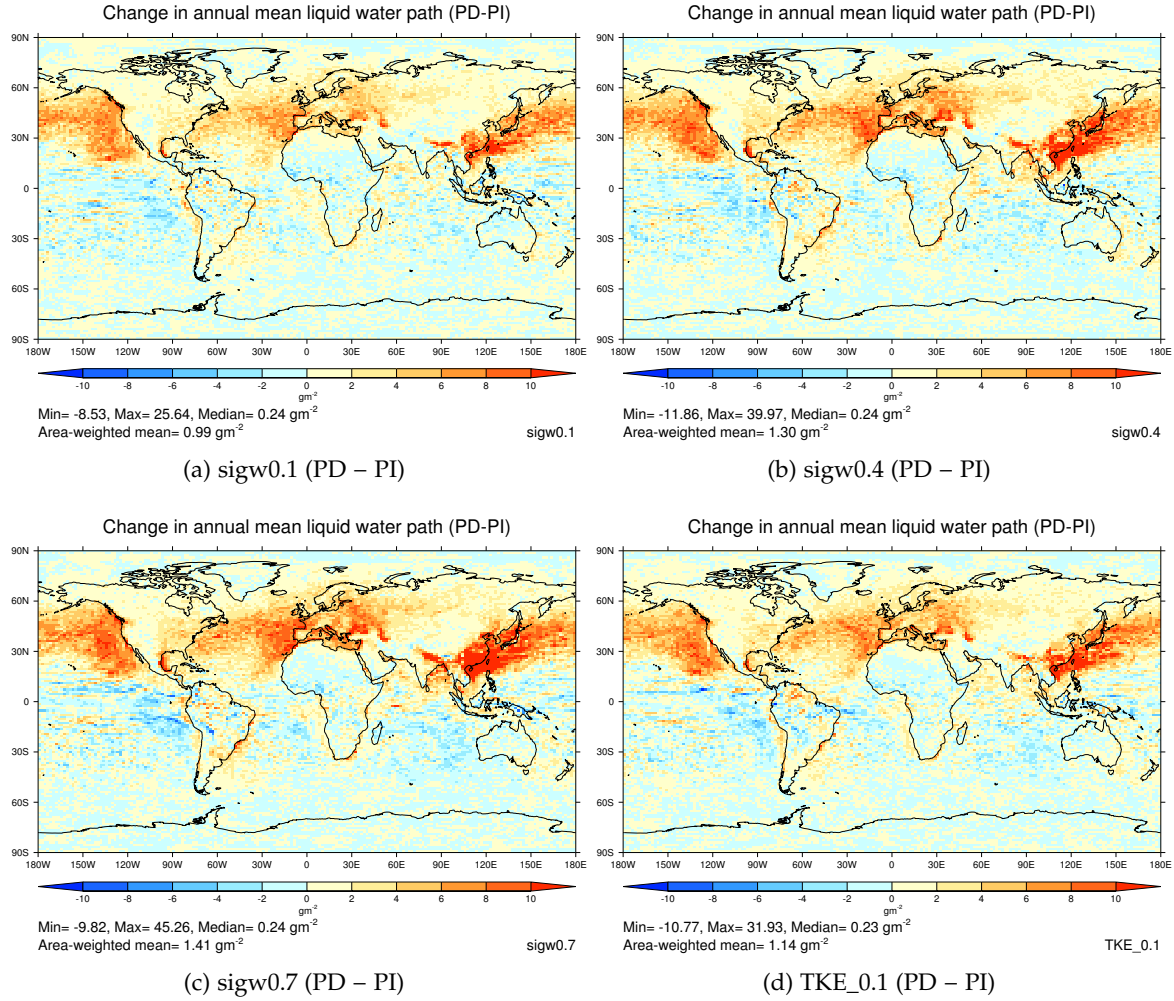


Figure 6.6: Absolute difference (PD - PI) in annual mean liquid water path for model runs (a) sigw0.1, (b) sigw0.4, (c) sigw0.7 and (d) TKE_0.1.

the TKE-derived σ_w have a spatially similar distribution to the other runs, with a magnitude slightly below those of sigw0.4.

6.2.1 Uncertainty due to vertical velocity parameterisation

The range of values of RFP generated by these configurations provides some quantification of uncertainty in estimates of the RFP due to the choice of parameterisation of vertical velocity. To illustrate how these four configurations compare to one another, Fig. 6.7 shows the zonal mean RFP due to anthropogenic aerosol effects for each of configurations sigw0.1, sigw0.4, sigw0.7 and TKE_0.1. The overall increasing magnitude of negative effects in the northern hemisphere with increasing σ_w is more clearly visible here than in the maps shown previously. The behaviour of TKE_0.1 is shown to be closer to sigw0.4 than sigw0.1, as expected from the global mean RFP. The near-zero zonal average RFP in the southern hemisphere masks significant regional variation. As discussed previously, while the RFP over land is mostly negative, over ocean in the southern hemisphere the effect is small but positive, with a region of elevated positive RFP in the southeastern Pacific. It appears

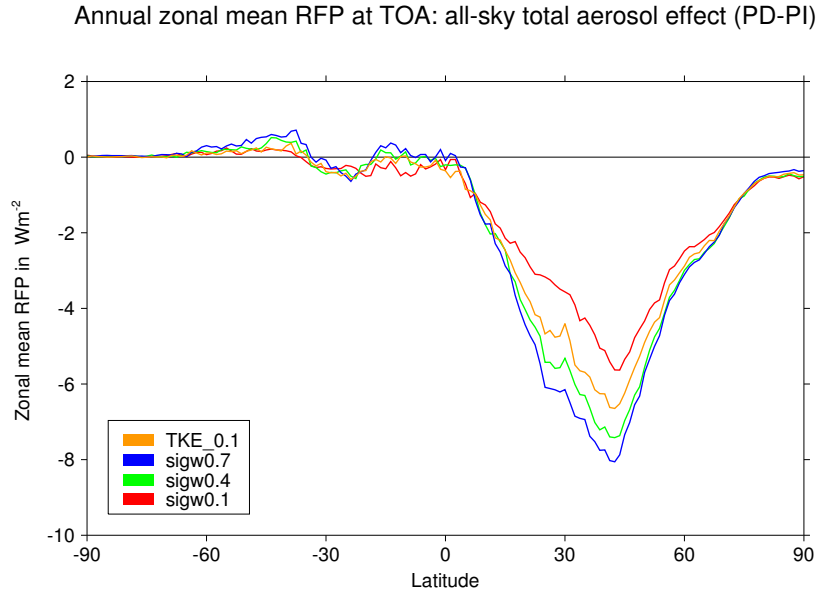


Figure 6.7: Annual zonal mean RFPs due to total anthropogenic aerosol effects for model configurations: sigw0.1 (red), sigw0.4 (green), sigw0.7 (blue), TKE_0.1 (orange).

that the magnitude of this positive effect also increases with increasing σ_w . This will be discussed in Sec. 6.3, below.

As described in Sec. 4.3, the choices of σ_w for sigw0.1 and sigw0.7 cover the range of the majority of the observed values of σ_w recorded in the 13 flight campaigns listed in the database, with very few exceptions. The limited spatial and temporal sampling of this set of observations notwithstanding, it is assumed that the range of values in the whole set is broadly representative of σ_w in boundary layer stratiform cloud. Should a fixed value of σ_w be chosen for a GCM, within the boundary layer, it should fall in the range $0.1 < \sigma_w < 0.7 \text{ m s}^{-1}$; although there may be individual cases where σ_w exceeds these values, to choose a fixed value of σ_w outside of this range to be applied globally would be grossly misrepresentative.

The annual mean RFPs from these two runs therefore provide an upper and lower bound due to the effect of the choice of updraught velocity parameterisation of estimates of the RFP in HadGEM-UKCA due to anthropogenic aerosols of -1.4 to -2.0 W m^{-2} . This range is likely to be more sensitive to the choice of bounds at the low end, because the sub-linear dependence of ΔCDNC (i.e. PD – PI) on σ_w saturates with increasing σ_w , and therefore it would be most beneficial to focus further study on forming a tighter constraint on the low end of the range.

Choice of σ_w within HadGEM-UKCA

As shown in Fig. 5.15, both TKE_0.1 and sigw0.4 generate CDNCs that compare reasonably well with those measured in marine stratiform clouds, which provides a degree of confidence in the estimates of the RFP. However, the comparisons against these 13 flight campaigns does not provide enough information to quantify whether sigw0.4 or TKE_0.1 is a better choice of vertical velocity parameterisation to be applied globally. In light of the temporal and spatial variability of σ_w shown with the flight campaigns, a parameterisation in which

σ_w also has the potential to vary is a more physically representative choice than one in which σ_w has a fixed value. However, the assumption that this variability can be meaningfully approximated by a Gaussian pdf with $\sigma_w = \sqrt{2/3\text{TKE}}$, is still open to further evaluation. In the meantime, TKE_0.1 could be used as the default setting for HadGEM-UKCA, with the recommendations that the issues of the high frequency of occurrence of $\sigma_w^{\min}$ (due to cloud base falling in between or just above the turbulent layers), and the choice of $\sigma_w^{\min}$ above the boundary layer, be addressed as a matter of urgency.

6.2.2 Splitting direct and indirect aerosol effects

Total anthropogenic aerosol effects can be conceptually divided up into the direct effects of aerosol on radiative balance and the indirect effects via the influence of aerosols on cloud. The coupling between these effects must also be taken into account since, if absorbing aerosols overlie clouds, their direct effect can be large (and positive) (Haywood and Shine, 1997). Secondly, clouds are modified by indirect aerosol effects, and these changes will in turn modify the direct effect of overlying aerosols. Thirdly, there is also a coupling term in clear-sky, where clouds can be prevented from forming via the semi-direct effect. The all-sky total RFP can be broken down into

$$\text{RFP(Total)}^{\text{allsky}} = (1 - f_c) \times \text{RFP(Direct)}^{\text{clearsky}} + f_c \times \left(\text{RFP(Direct)}^{\text{cloudysky}} + \text{RFP(Indirect)}^{\text{cloudysky}} + \text{coupling} \right), \quad (6.2)$$

where f_c is the cloud fraction. The semi-direct effect is very difficult to incorporate into this framework, due to the modification to f_c . In order to attempt to disentangle the direct effects from the total, a second pair of simulations was run with direct effects only, i.e. indirect aerosol effects are switched off (i.e. fixed values of N_d are used in the calculations of effective radius [Eq. (3.2)] and autoconversion [Eq. (3.4)]). Fixing the values of CDNC in this second pair of runs means that the reference climate is necessarily different between the two pairs of runs, and so this method can only give approximate figures for the split between direct and indirect effects out of the total aerosol RFP. We then make the assumption that the indirect effects (and couplings) can be estimated by subtracting the direct effects from the total, i.e.

$$\text{RFP(Indirect)}^{\text{allsky}} = \text{RFP(Total)}^{\text{allsky}} - \text{RFP(Direct)}^{\text{allsky}}. \quad (6.3)$$

(It is impossible to extract the coupling term, so for these purposes it is considered to be a contribution to the indirect effects.)

The first three maps in Fig. 6.8 show the radiative effects in model TKE_0.1 split up in this way: (a) shows the RFP due to total aerosol effects; (b) shows the RFP from a pair of runs with the direct effect only; and (c) shows the results of subtracting (b) from (a), which provides an estimate of the contributions to the total RFP from the indirect effects. The direct effects are found to have a negative effect on average of -0.6 W m^{-2} , and weaker than the indirect effects, with a noisy signal over ocean. The dominant contribution to the total aerosol effects comes from the indirect effects (-1.1 W m^{-2} out of a total of -1.7 W m^{-2}).

These indirect effects can be split up further to provide separate estimates of contributions from the cloud albedo effect and cloud lifetime effects by a similar method. Two further pairs of runs of model TKE_0.1 were carried out, 'NoAlb' and 'NoLife'. In 'NoAlb' the cloud albedo effect was switched off by using a fixed value of CDNC for the effective radius calculation, Eq. (3.2). The cloud albedo effect itself can then be estimated

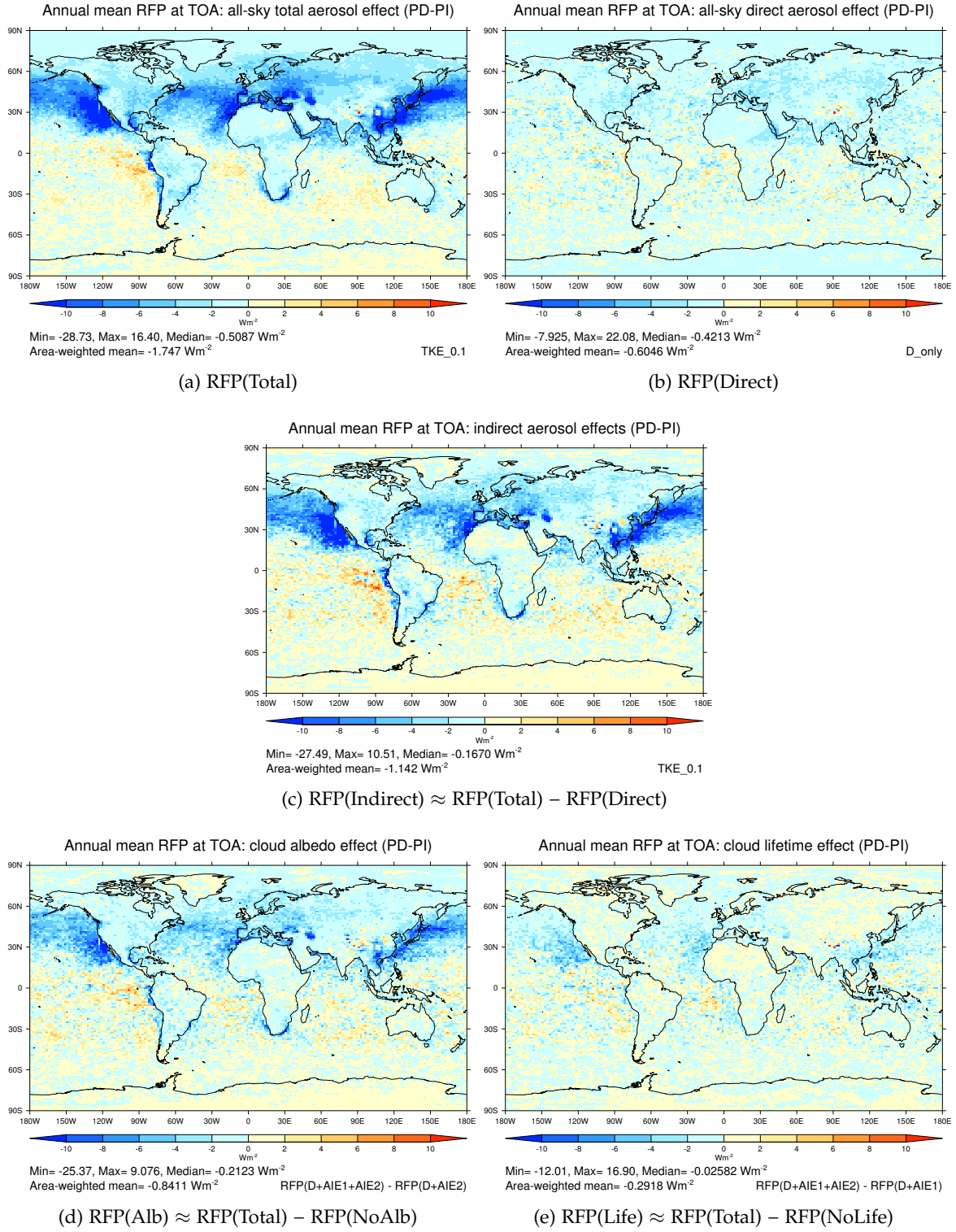


Figure 6.8: Annual mean radiative flux perturbations calculated from model TKE_0.1, run with the following combinations of radiative effects: (a) Total aerosol effects (both direct and indirect effects); (b) Direct effects only (indirect effects are switched off); (c) Indirect effects, inferred by subtracting (b) from (a); (d) Cloud albedo effect, inferred by subtracting the RFP from a pair of runs with direct and cloud lifetime effects only, from the total RFP (a); (e) Similar to (d), i.e. cloud lifetime effect, inferred by subtracting the RFP from a pair of runs with direct and cloud albedo effects only from the total RFP (a). Note that inferred fields ignore couplings between effects.

using $\text{RFP}(\text{Alb}) \approx \text{RFP}(\text{Total}) - \text{RFP}(\text{NoAlb})$. Similarly, the cloud lifetime effects were switched off in the pair of runs 'NoLife', by using a fixed value of CDNC in the autoconversion rate equation, Eq. (3.4), to allow the cloud lifetime effects to be estimated from $\text{RFP}(\text{Life}) \approx \text{RFP}(\text{Total}) - \text{RFP}(\text{NoLife})$. Maps of these effects are shown in (d) and (e) of Fig. 6.8. Again, using fixed values of CDNC in either the effective radius or autoconversion rate equations respectively means that the reference climate for the pair of runs 'NoAlb' is not identical to that for 'NoLife', but, to the extent that it is legitimate to split up these highly coupled effects in this way, the cloud albedo effect (-0.84 W m^{-2}) is found to be stronger than the cloud lifetime effects (-0.30 W m^{-2}), while the spatial distribution of both effects is similar. However, cloud lifetime effects have been shown to be strongly dependent on the choice of autoconversion parameterisation (Golaz et al., 2011), so this number is subject to change should the autoconversion process become better constrained in future.

Interestingly, it is found that the sum of the RFPs due to each of the aerosol effects considered in isolation (cloud albedo, cloud lifetime, direct effect), each calculated from independent pairs of runs, comes to -1.74 W m^{-2} , which is close to the estimate of the RFP of the total aerosol effect, of -1.74 W m^{-2} (obtained from one pair of runs that includes all of the effects), indicating that in HadGEM-UKCA, the individual effects are nearly additive, which is a noteworthy result (Haywood et al., 2009).

6.3 Discussion

A slightly surprising feature of the RFP maps shown in Fig. 6.2 is the sign difference in the effect between the northern and southern hemispheres, and in particular the region of elevated positive forcing in the southeastern Pacific. A possible explanation for this is found by considering the microphysical aerosol processes at work in UKCA.

6.3.1 Aerosol distributions

As described in Chapter 3, the configuration of GLOMAP-mode used for the work in this thesis consists of an external mixture of five aerosol modes, composed of internal mixtures of prognostic sea salt, anthropogenic black carbon, organic carbon and sulphate. Mineral dust is transported separately.

Mass

The annual mean aerosol burden (column-integrated mass) of each of these components simulated by GLOMAP-mode (i.e. not dust) is shown in Figs. 6.9 and 6.10. Results are shown for pairs of runs of the reference model (sigw0.4) and illustrate the difference in resulting aerosol burden of each component between pre-industrial and present day aerosol emissions. Relative changes in burden are shown alongside absolute differences between present day and pre-industrial aerosol.

On average, the sulphate burden has tripled between the PI and PD runs, but the maps show that the increase is far stronger in the northern hemisphere (ten-fold increases in some regions) than the southern (typical increases of 40 to 80%). Over land, increases are particularly strong in the industrialised and heavily populated regions of China, India, the Middle East, eastern North America and much of Eurasia. Sulphate burden has also at least doubled over most of the ocean in the northern hemisphere and along the major shipping routes through the southern hemisphere. This large increase in the mass of sulphate in the northern hemisphere is a major contributing factor to the strong negative RFP seen in

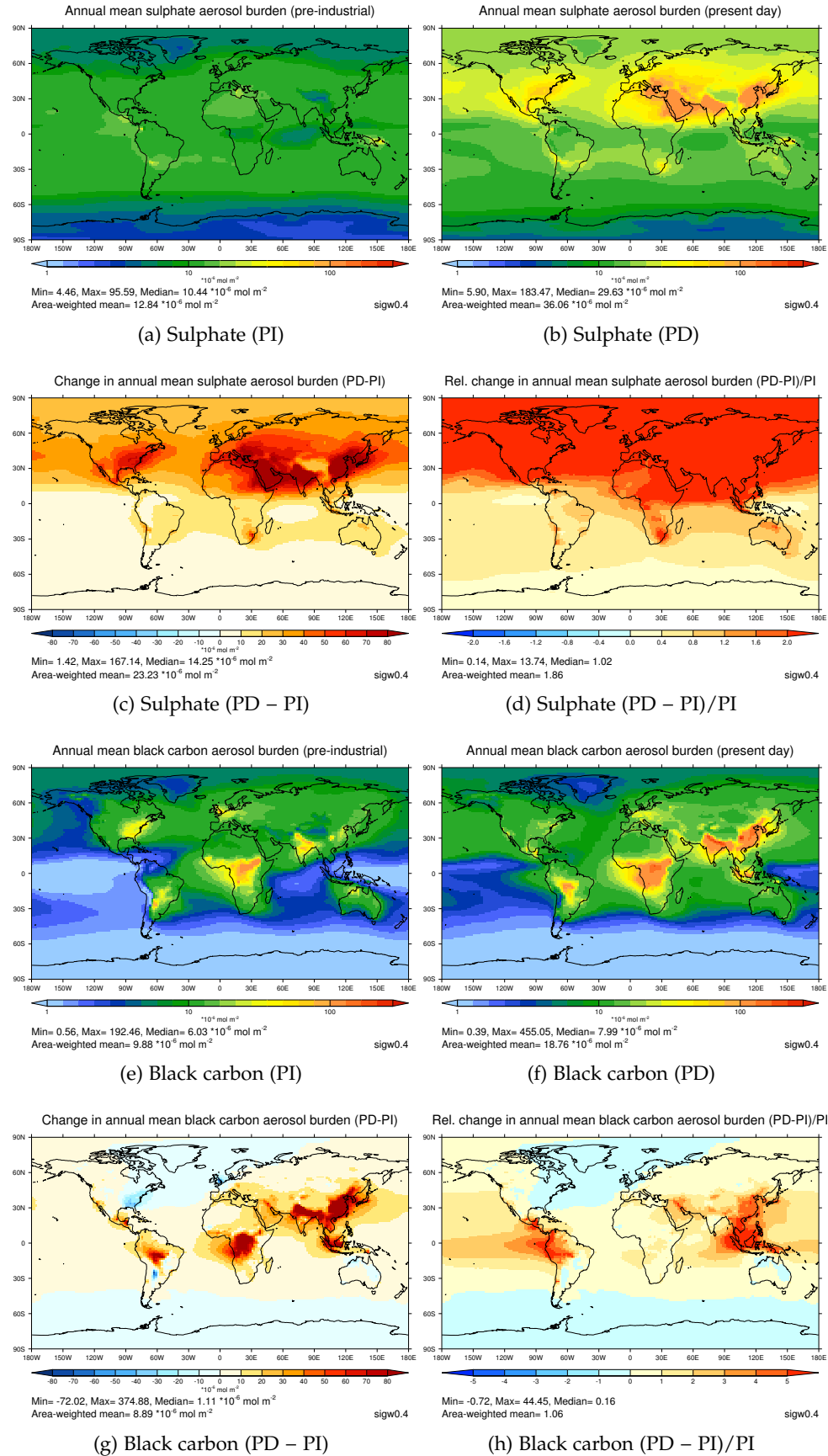


Figure 6.9: Annual mean aerosol burden by component: (a–d) sulphate, (e–h) black carbon. All results from model sigw0.4 run with: (a) & (e) pre-industrial aerosol emissions, (b) & (f) present day aerosol emissions; (c) & (g) show absolute differences (PD – PI); (d) & (h) show relative differences (PD – PI)/PI.

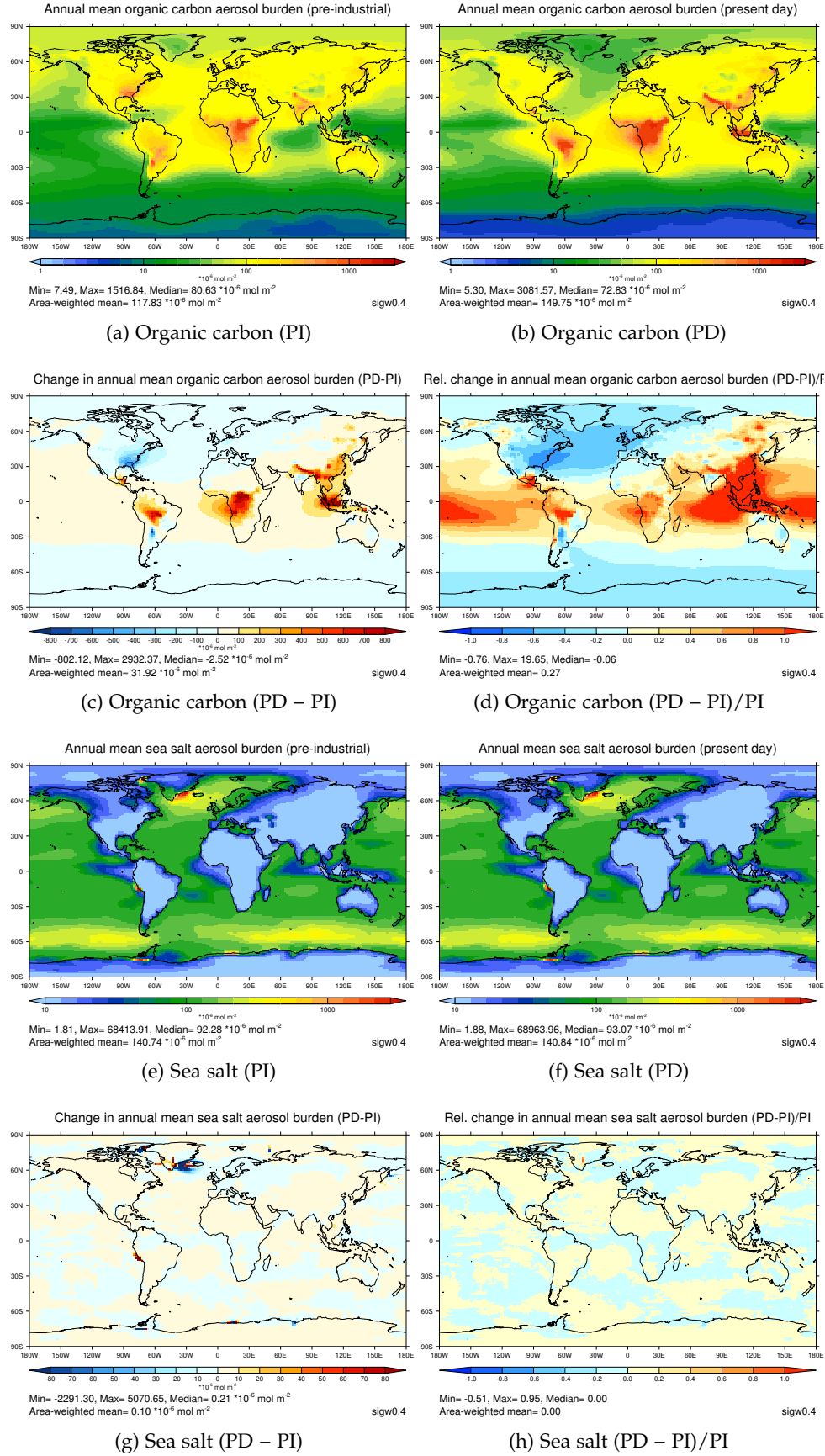


Figure 6.10: Continuation of Fig. 6.9. Annual mean aerosol burden by component: (a–d) organic carbon and (e–h) sea salt.

the northern hemisphere, due to the increase in CCN, although exact details are of course dependent on the size and number distribution of this increased mass. The sharp contrast between the hemispheres seen in the difference plots of sulphate burden is clearly followed through to the contrast seen between the same regions in the RFP.

Increases in the black carbon burden due to a combination of fossil fuel combustion and biomass burning has produced a doubling of the global average burden, and substantially greater increases in localised regions. Significant relative increases have occurred over China and Indonesia, and over the Amazon, sweeping out across the tropical Pacific. Over eastern North America, the north Atlantic and northern Europe, the black carbon burden has slightly decreased. The global mean burden of organic carbon has increased 25% between the PI and PD runs, with strong increases in the biomass burning regions and over China. Similar to black carbon, 40 to 60% decreases have occurred over North America, the North Atlantic and northern Europe. The sea salt burden has not changed between the two runs.

Number

While the difference in total mass of each component is mainly determined by the differences between the pre-industrial and present day emissions, the number and size distribution of particles is controlled by the aerosol and chemical processes that are explicitly modelled by UKCA. How this mass is distributed in terms of particle size and numbers can significantly affect the CDNC and hence the indirect aerosol effects.

6.3.2 Influence of aerosol microphysics on radiative flux perturbations

In the maps of RFP shown in Fig. 6.2, a weak positive effect is visible over the tropical oceans and the oceans of the southern hemisphere. As shown in Fig. 6.8, this positive region still remains once the direct aerosol effects have been subtracted out from the total effects, and must therefore be due to indirect aerosol effects. This positive effect is strongest in a region of the southeastern Pacific, and is seen to increase in intensity with increasing σ_w .

The cloud fraction in this region does not change significantly between the PI and PD runs (not shown). The effect must therefore be brought about by the difference between cloud droplet number concentrations in runs with PI and PD aerosol emissions. Indeed, as shown in Fig. 6.4, the number of cloud droplets has decreased in the PD run compared to PI, in the region of positive RFP.

To confirm the existence of changes in cloud-relevant aerosol number, Fig. 6.11 shows maps of CCN at fixed supersaturation for runs of sigw0.4 with pre-industrial and present day aerosol emissions, and the difference between these. (A high supersaturation of $S = 1\%$ is chosen to emphasise contrasts.) The difference plot shows a decrease in the number of CCN in the relevant region, indicating that the number of CDNC decreases because there are fewer activatable aerosols in the region.

The response of the RFP in fixed σ_w runs provides further information. We have shown that CDNC increases with increasing σ_w , because increasing σ_w can increase $S_{\max}$, which causes $a_{\min}$, the dry radius of the smallest particle which activates, to be lower, which means that more of the smaller particles can activate in the higher updraughts. Further, the magnitude of ΔCDNC between PD and PI—both positive and negative—increases with increasing σ_w (shown in Figs. 6.4 and 6.5). This could be the result of a number of different effects: a simple increase in the total number of soluble aerosols, while the distribution kept the same shape, or more complex changes to the distribution of size, number, mass, or composition between PI and PD runs, as a result of microphysical aerosol interaction.

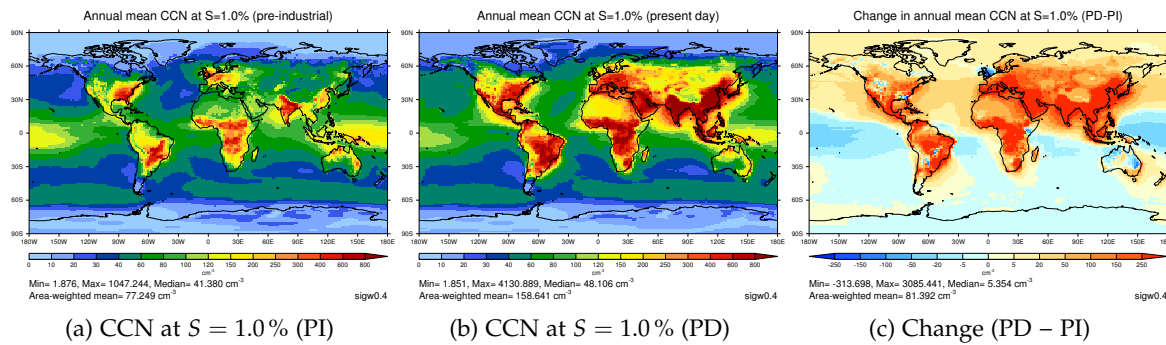


Figure 6.11: Annual mean CCN at $S = 1.0\%$ in model sigw0.4 run with (a) pre-industrial and (b) present day aerosol emissions; (c) shows difference (PD – PI).

So, is the number of small, soluble aerosols lower in the PD than PI runs in this region? And if so, why? Figs. 6.12, 6.13 and 6.14 show the annual mean number burden (column-integrated number concentration) in each of the five size modes used in this configuration of UKCA, the four soluble aerosol modes (nucleation, Aitken, accumulation and coarse) and the insoluble Aitken mode. The possible constituent components of each of these modes is listed in Table 3.1. For each mode, maps of pre-industrial and present day annual mean number burden are presented, as well as the absolute change (PD – PI), and the relative change (PD – PI)/PI.

Fig. 6.12 presents the number burden in the two smallest modes, and both nucleation and Aitken modes show a decrease in the number of particles in the relevant region from PI to PD runs of between 20 and 40%. It is evident that despite increased SO_2 emissions (not shown, but implied by increase in sulphate burden shown previously in Fig. 6.9), fewer new sulphate particles nucleate in PD conditions, and there are subsequently fewer sulphate particles that can grow into the Aitken mode. The absolute decrease of Aitken mode particles is particularly strong across the tropical Pacific, which corresponds to the region of elevated positive RFP seen in figures Fig. 6.2.

By contrast, the number burden of aerosols in the soluble accumulation mode has increased almost universally in the PD compared to PI run, as shown in Fig. 6.13, and has doubled on average. Strong increases (up to eight-fold) are notable in both biomass burning and industrial regions, and relative increases of between 60 and 80% are also visible in more remote regions, such as across the southern Pacific.

The number of particles in the coarse soluble mode is dominated by sea salt, and shows very little change between runs with pre-industrial and present day aerosol emissions, as shown in Fig. 6.13.

Number burdens of the insoluble Aitken mode are shown in Fig. 6.14. Emissions of insoluble particles in the industrial and biomass burning regions increase between PI and PD emissions, which leads to the strong increase seen across south-east Asia, the Indian Ocean and the tropical Pacific. Although insoluble aerosols cannot themselves act as CCN, interactions with soluble components lead to indirect influences, e.g. via coagulation with soluble particles and condensation of soluble components.

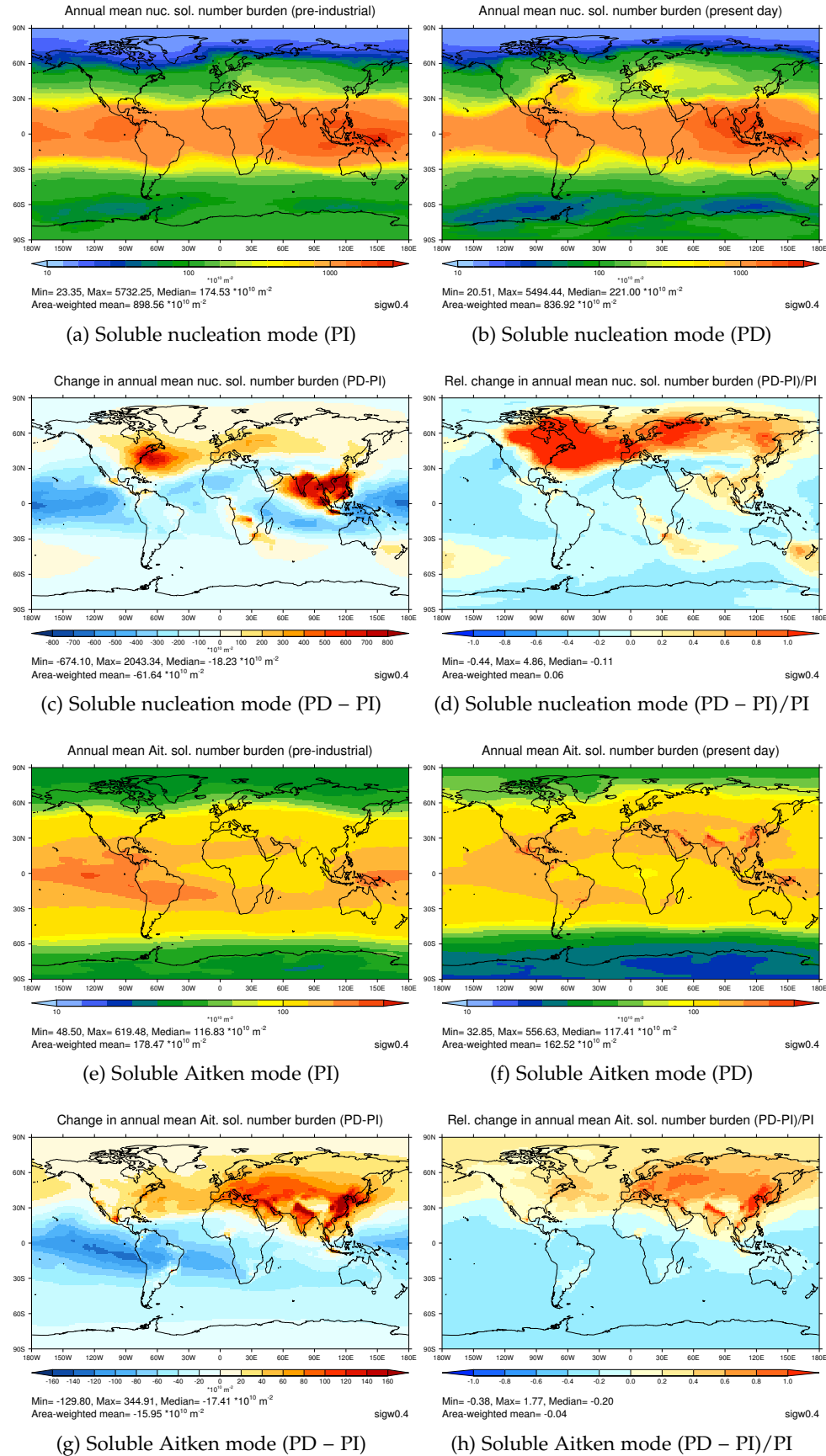


Figure 6.12: Annual mean aerosol number burden by mode: (a–d) soluble nucleation mode, (e–h) soluble Aitken mode. All results from model sigw0.4 run with: (a) & (e) pre-industrial aerosol emissions, (b) & (f) present day aerosol emissions; (c) & (g) show absolute differences (PD – PI); (d) & (h) show relative differences (PD – PI)/PI.

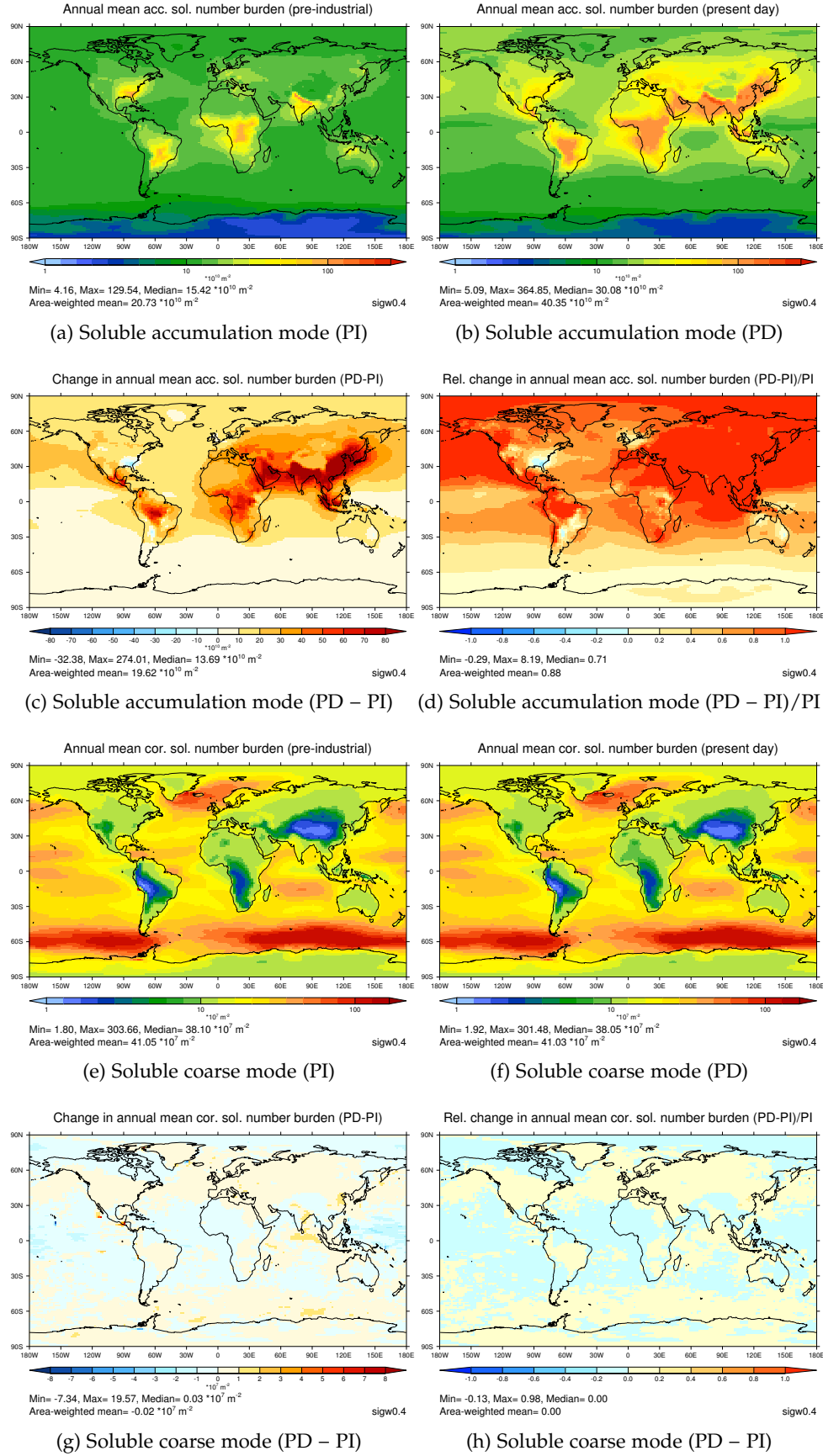


Figure 6.13: Continuation of Fig. 6.12. Annual mean aerosol number burden by mode: (a–d) soluble accumulation mode, (e–h) soluble coarse mode.

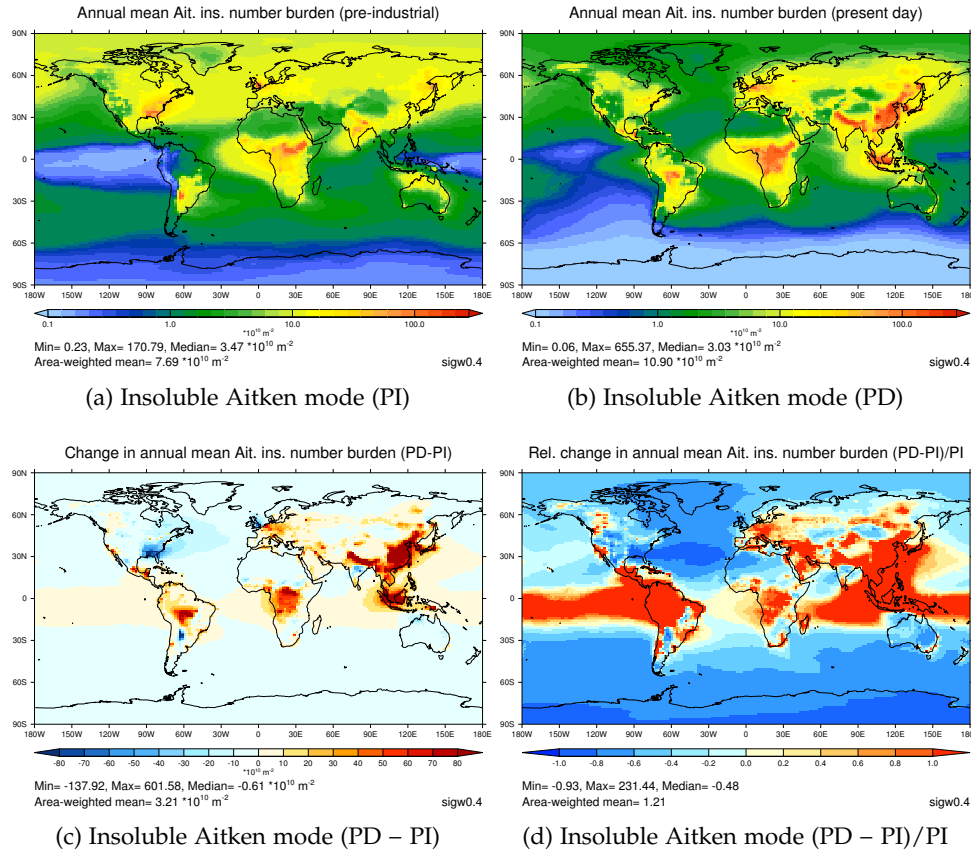


Figure 6.14: Continuation of Fig. 6.12 and Fig. 6.13. Annual mean aerosol number burden in insoluble Aitken mode.

Suppression of nucleation

A possible explanation for the suppression of nucleation of fresh sulphate particles and subsequent reduction in the number of CCN-relevant soluble Aitken mode particles in the southeastern Pacific is that the increased number of soluble accumulation and insoluble Aitken mode particles in this previously-pristine region provides an increased surface area on to which H_2SO_4 vapour can condense. This process of condensation on to existing particles is more energetically favourable than new particle formation, and thus occurs preferentially, effectively shutting off a large fraction of the particle nucleation.

Conversely, the number of nucleation mode particles has a strong relative increase over North America, the north Atlantic and Northern Europe potentially due to the inverse of the same process, as these are regions where the number of insoluble Aitken particles decreases.

However, to complicate matters, in regions such as south-east Asia, nucleation of sulphate particles increases substantially despite an increase in both accumulation and insoluble Aitken mode numbers. This is possibly because the absolute abundance of SO_2 emitted in this region in the PD run is so large, that, following oxidation, sulphate aerosol nucleation occurs regardless of the additional condensational sink provided by additional larger particles.

The complex interplay between the microphysical aerosol processes represented within

UKCA, combined with the highly non-linear aerosol activation scheme, leads to responses to perturbations that would be impossible to predict from physical intuition alone. These responses are shown to play out in the climate-relevant radiative effects, which underlines the importance of the inclusion of microphysical aerosol modelling in GCMs. Essentially, microphysics matters!

6.4 Comparison with other estimates of aerosol radiative effects

Current estimates of the radiative effects of anthropogenic aerosol are associated with large uncertainties. The most recent Intergovernmental Panel on Climate Change (IPCC) Assessment Report (Forster et al., 2007) estimates the direct aerosol radiative forcing to be $-0.5 \pm 0.4 \text{ W m}^{-2}$. This large uncertainty range is primarily due to differences between modelling and observation-based estimates. A more recent study incorporating the results of a global aerosol model adjusted to an observation-based method estimates the direct aerosol forcing to actually be weaker than this, with a best estimate of -0.3 W m^{-2} (Myhre, 2009). This is half the magnitude of the HadGEM-UKCA estimate of -0.6 W m^{-2} , so it is possible that the direct effect of aerosols is too strong in UKCA.

Of the indirect aerosol effects, only the cloud albedo effect may be calculated as a strict radiative forcing, and the IPCC provide a best estimate of -0.7 W m^{-2} , with a 5 to 95% confidence range -0.3 to -1.8 W m^{-2} . Although it is not strictly valid to compare RFPs directly with radiative forcings, the estimate from UKCA-Activate of the contribution to the total RFP from the cloud albedo effect is -0.8 W m^{-2} (with configuration TKE_0.1), which is within the range of radiative forcings reported by the IPCC.

The IPCC report also includes a review of estimates of RFPs due to the total aerosol effect. This varies widely from -0.2 W m^{-2} in a set of combined GCM and satellite simulations (Quaas et al., 2006) to -2.3 W m^{-2} in other GCM simulations (Ming et al., 2005), with an average forcing of -1.2 W m^{-2} (Denman et al., 2007). However, other than the fact that there are clearly large uncertainties associated with aerosol effects, it is difficult to draw further conclusions from this wide range of results. Even just considering the modelling studies alone, there are many differences between the simulations in terms of both how the aerosols are modelled (and indeed, which aerosols are included) and the parameterisations of cloud processes. Models vary considerably in their representation of cloud droplet activation, cloud droplet spectral dispersion, autoconversion of cloud water to rain water, which have all been shown to introduce significant uncertainties to estimates of radiative effects (Chen and Penner, 2005). The cloud lifetime effect is particularly sensitive to the parameterisation of autoconversion (e.g. Jones et al. (2001); Menon et al. (2002, 2003); Penner et al. (2006)). In fact, in one model the total aerosol RFP has been found to be directly proportional to the autoconversion threshold (Golaz et al., 2011) indicating the clear imperative to tighten the constraint on this process for future modelling studies.

A number of model intercomparison studies since the publication of the IPCC Fourth Assessment Report have investigated what causes the wide range of model estimates of indirect aerosol effects. The AeroCOM initiative has established a framework with which to do this, starting with a unified set of prescribed aerosol emission inventories which have been used by participating GCMs in several carefully-designed experiments. For example, Penner et al. (2006) examined the differences in behaviour of three GCMs in a series of experiments, starting with a tightly controlled experiment in which aerosol concentrations, CDNC parameterisation and autoconversion scheme are specified, to one in which only the aerosol sources are prescribed. This study found that large differences in the indirect

aerosol effects were attributable to choice of autoconversion parameterisation but that the largest differences were due to the modelling of aerosols themselves. Given a fixed set of aerosol sources, the diversity in the eventual prediction of aerosol concentrations led to values of cloud forcing ranging from -0.3 W m^{-2} to -1.4 W m^{-2} .

In a later AeroCOM intercomparison study, Quaas et al. (2009) compared the statistical relationships between several cloud properties and aerosol optical depth in ten GCMs and compared these with the relationships found in satellite data. The strength of these relationships in both model and satellite data was combined to calculate a global mean shortwave total aerosol forcing estimate of $-1.5 \pm 0.5 \text{ W m}^{-2}$, in which the statistical uncertainty is derived from the combination of models and satellite observations. The wide range of estimates of the total aerosol RFP found with UKCA, from -1.4 W m^{-2} to -2.0 W m^{-2} , falls just within this range.

The range of the mean responses of different models in model intercomparison studies highlights some of the structural uncertainties in modelling of the indirect aerosol effects, due to differences in the way that both aerosol and clouds are modelled in participating GCMs, and how they are allowed to interact. The overall uncertainty is also due to uncertainties in emissions scenarios and parametric uncertainties, that is, the range of results obtained by varying a single parameter, within plausible bounds. Two recent studies have focused on the impact of different cloud-related parametric uncertainties on anthropogenic aerosol effects. Lohmann and Ferrachat (2010) found that tuning the present day climate to be in radiative balance does not seem to have an influence on estimates of the total anthropogenic aerosol effect. Four cloud-related parameters commonly used for tuning the present day climate were investigated, including the autoconversion rate. They found a very small difference in the total anthropogenic effect of aerosols between runs where the climate was in radiative balance and those when it was not. They also found that even when varying all four of these parameters over the full ranges explored, the anthropogenic aerosol effect in their model only varied by 25% (or 11% for the in-balance runs). However, in a study with a different model, Golaz et al. (2011) found that the amount the autoconversion threshold needed to be retuned in order to obtain radiative balance in runs with different parameterisations led to an RFP that was proportional to the autoconversion threshold itself, and that varies by $\pm 50\%$ (from -0.42 to -1.41 W m^{-2}). This leads to the conclusion that parametric uncertainties can vary between models due to the different structural configuration of the model in question, or due to the interactions between parameterisations, and that although knowledge of the parametric uncertainty in one model is important in understanding how that model responds to particular parameters, it would not necessarily translate identically to other models.

6.4.1 Previous estimates of aerosol effects in the UM

Prior to the introduction of UKCA, recent estimates of the radiative effects of aerosols within the Unified Model have used a mass-based aerosol scheme (known as CLASSIC, the Coupled Large-scale Aerosol Simulator for Studies in Climate, summarised in Bellouin et al. (2011)) with an empirical relationship for cloud droplet number concentration (Jones et al., 2001). One of the key differences between the UKCA and CLASSIC aerosol schemes is that UKCA simulates not only aerosol mass, but also aerosol number (Mann et al., 2010). This means that processes that affect aerosol mass without affecting aerosol number, and *vice versa*, can both be properly represented. This is important when representing particle nucleation, the simulation of CCN, and hence the indirect aerosol effects (Merikanto et al., 2009).

The main difference between the implementation of the Jones et al. 2001 empirical relationship for CDNC between simulations with the CLASSIC aerosol scheme and those with aerosol from UKCA in configuration JonesCDNC, is that in CLASSIC, CDNC is calculated by taking the simulated aerosol mass and then calculating the number of aerosols, assuming that that mass is organised into a fixed size distribution. Thus, positive RFPs could only occur in regions where there were a decrease in aerosol mass between PI and PD conditions. However, in UKCA, the aerosol size distribution varies dynamically. For this reason, positive values of RFP can be obtained due to a change in the distribution of aerosol number in the modes, even without a decrease in the soluble aerosol mass.

The most recent published estimates of radiative forcing from the CLASSIC aerosol scheme running in the Hadley Centre Earth System model, HadGEM2-ES, (Collins et al., 2011) are a direct forcing of -0.2 W m^{-2} and a direct plus cloud albedo effect forcing of -1.5 W m^{-2} from a fully coupled run with CMIP5 emissions between 1860 and 2000 (Bellouin et al., 2011). The (unpublished) RFP from this model setup, with direct and cloud albedo effects only, was calculated as -1.4 W m^{-2} .

The estimate from HadGEM-UKCA in configuration TKE_0.1 of the RFP due to the direct and cloud albedo effect (experiment NoLife) is found also to be -1.4 W m^{-2} although the RFP due to the direct effect in HadGEM-UKCA is a greater fraction of this (-0.6 W m^{-2} out of -1.4 W m^{-2}) than the radiative forcings found with CLASSIC (-0.2 W m^{-2} out of -1.5 W m^{-2}). The estimate of the cloud albedo effect from CLASSIC and the direct effect from UKCA are found to be on the high side of the IPCC AR4 ranges. No estimate for the RFP due to cloud lifetime effects was made in these CMIP5 runs with CLASSIC aerosol, but a previous estimate of this effect from the Unified Model of -0.5 W m^{-2} (Jones et al., 2001), is greater than the estimate of -0.3 W m^{-2} made with configuration TKE_0.1 of UKCA.

The total aerosol RFP from HadGEM-UKCA in configuration JonesCDNC is -2.3 W m^{-2} , which gives an estimate of the indirect aerosol effects of -1.7 W m^{-2} , once the RFP due to direct effects has been subtracted out. This is smaller than a previous estimate of RFP due to the indirect effects of sulphate of -1.9 W m^{-2} (Jones et al., 2001).

6.5 Conclusions

This chapter has presented the radiative flux perturbations due to total anthropogenic aerosol effects in six different configurations of UKCA-Activate, and placed them in the context of other recent estimates of these effects.

In the different configurations of vertical velocity, Fig. 6.1 illustrates how increasing the standard deviation of a Gaussian pdf of updraught globally from $\sigma_w = 0.1 \text{ m s}^{-1}$ to 0.7 m s^{-1} generates RFPs of total anthropogenic aerosol effects ranging from -1.4 W m^{-2} to -2.0 W m^{-2} . This range is a fairly significant fraction of the total aerosol forcing estimate of $-1.5 \pm 0.5 \text{ W m}^{-2}$ found in the Quaas et al. (2009) AeroCOM intercomparison study, involving ten GCMs and relationships derived from satellite data. This demonstrates the importance of using an empirically-verified choice of updraught velocity parameterisation when modelling indirect aerosol effects.

The magnitude of RFP increases with σ_w , but this effect saturates locally once all potential CCN have activated. The uncertainty due to this parameter alone is estimated to be about a third of the overall total anthropogenic aerosol effect. While the range of σ_w explored here extends to the extremes of what would be a physically reasonable approximation to apply globally for stratiform clouds, it demonstrates how the choice of one parameter in the vertical velocity parameterisation can make a significant impact on

the RFP estimate of a model.

Examining the difference between the maps of RFP shows where this choice can make the most difference. Fig. 6.3(a) shows a map of the difference between the RFP generated by sigw0.7 compared to sigw0.1. Outstanding features highlight the regions where increasing σ_w has the greatest effect on CDNC that is most relevant to the RFP, i.e. frequently cloudy regions with high CCN concentrations. In these regions, it is particularly important to use a realistic pdf of updraught velocities, since these are the regions in which the RFP is most susceptible to the presence of stronger updraughts. This information could inform the location of future flight campaigns.

Most flight campaigns studying aerosol–cloud interactions have focused on marine stratocumulus clouds, where the CDNC tends to be limited by the number of CCN, rather than the updraught velocity. Of course, it is therefore of great importance to have detailed and accurate measurements of CCN in these regions. However, in terms of vertical velocity measurement, Fig. 6.3(a) indicates that inaccuracies in choice of σ_w for a GCM can have the most impact in other frequently cloudy regions with high CCN concentrations, particularly in the industrialised and heavily polluted regions of the northern hemisphere, such as China and East Asia, the Ganges plain and, to a lesser extent, the whole of eastern Europe. For the purposes of reducing the uncertainty in estimates of the indirect aerosol effects due to the parameterisation of vertical velocity it would be particularly useful to have more measurements of the vertical velocity distributions in continental stratiform and shallow cumulus clouds in these regions.

In terms of the best estimate of RFP from UKCA-Activate, there is insufficient evidence to make the choice between TKE_0.1 (RFP = -1.7 W m^{-2}) and sigw0.4 (RFP = -1.9 W m^{-2}). While the spatial and temporal variability of σ_w found in TKE_0.1 is more realistic than applying a fixed value of $\sigma_w = 0.4 \text{ m s}^{-1}$ everywhere, technical restrictions within the current configuration of HadGEM-UKCA (such as the absence of a TKE diagnostic above the planetary boundary layer) causes an unrealistically high frequency of occurrence of the minimum value in this configuration.

In the spatial differences between the RFPs of TKE_0.1 and sigw0.4 shown in Fig. 6.3 (b), attention is drawn to two points. First, as for the difference between sigw0.7 and sigw0.1, the largest ΔRFP is seen in a region of high CCN concentration over China. In this region the TKE-derived σ_w obtained with TKE_0.1 is usually below 0.4 m s^{-1} , hence fewer aerosols activate and the indirect aerosol effects are weaker than in sigw0.4. Measurements of σ_w in this region would be helpful in choosing between these two model configurations.

The second point relates to the absence of a notable ΔRFP signal in the persistent marine stratocumulus regions. In Sec. 4.4 the highest values of σ_w in TKE_0.1 were found to be in the stratocumulus regions off the west coasts of North and South America. However, there is little change in the RFP in these regions between the TKE_0.1 and sigw0.4 models because a high fraction of activation can be obtained with relatively low updraughts, i.e. here, it is the number of potential CCN which is the limiting factor on CDNC. Thus there are several climatically-important regions where accuracy in aerosol modelling can have a greater impact on the indirect aerosol effects than the parameterisation of the activation of those aerosols to cloud droplets.

In fact, closer scrutiny of the differences in column-integrated aerosol number and mass in Figs. 6.9 to 6.14 shows that the aerosol microphysics can result in somewhat unexpected behaviour under a present-day aerosol emissions scenario compared to pre-industrial, particularly in remote regions. In pre-industrial conditions, the main source of aerosol in these regions is due to the production of new particles via the nucleation of sulphuric acid vapour. However, under present-day aerosol emissions, the increased number

of soluble accumulation and insoluble Aitken mode particles in the vicinity provides a large condensational sink for the H_2SO_4 vapour, and effectively shuts off the new particle nucleation process. This in turn results in a reduction of the number of CCN, compared to pre-industrial times and hence the sign of the indirect aerosol effects are seen to be reversed in these regions, resulting in areas of weakly positive RFP.

The modelling of indirect aerosol effects is dependent on accurate CCN concentrations, which are a result of the interactions of several highly non-linear aerosol microphysical processes. Thus responses to differing aerosol emissions scenarios are difficult to predict without a detailed model of these processes. What fraction of these potential CCN then actually activate to become cloud droplets is further dependent on the highly non-linear parameterisation of aerosol activation. The response of the climate to this combination of non-linearities underpinning aerosol–cloud interactions is beginning to be understood through the coupling of such microphysical aerosol and cloud processes with dynamical processes such as turbulence and updraught velocity in GCMs.

Conclusions

The indirect radiative effects of aerosols are currently the most uncertain of all known perturbations to the Earth's radiative balance. Reducing this uncertainty is therefore of critical importance for future climate predictions, which have significant implications for international climate policy. The work in the preceding chapters has attempted to provide some insight into this uncertainty with the aid of computational climate simulations with a general circulation model, HadGEM-UKCA.

A particular focus of the work has been on quantifying the uncertainty in the modelled indirect aerosol effects due to the choice of parameterisation of sub-grid-scale variability of updraught velocity required by the aerosol activation parameterisations used for large-scale cloud in global climate models.

Chapter 1 introduced the physics of aerosol activation and its parameterisation. Chapter 2 then evaluated two specific parameterisations and Chapter 3 described the implementation of one of these into HadGEM-UKCA. The work in Chapters 4 to 6 focused on understanding the effects of different vertical velocity configurations on modelled cloud properties in HadGEM-UKCA (Chapter 4), evaluating these against observations from flight campaigns (Chapter 5), and ultimately, on quantifying the impact on estimates of the indirect aerosol effects (Chapter 6). More detailed summaries of each of these chapters are provided below, followed by general conclusions and, in light of these, some recommendations for future work.

7.1 Summary and conclusions by chapter

Chapter 1

The first chapter provided an overview of the motivation and methodology for modelling the indirect aerosol effects with a global climate model. The activation of aerosols to cloud droplets is a microphysical process which must be parameterised in order for the large-scale effects to be incorporated into climate simulations. The historic development of aerosol activation parameterisations was presented, since this is the foundation for the work on parameterisation development covered in Chapter 2.

Chapter 2

The second chapter focused on two contrasting Köhler-theory based parameterisations of aerosol activation, one proposed by Abdul-Razzak and Ghan (2000) (referred to as

ARG) and the other devised by Fountoukis and Nenes (2005) (FN). The chapter presented a comparison of the two methods, and of the behaviour of both schemes in controlled box-model simulations.

These tests demonstrated some unexpected behaviour of the FN scheme in a region of parameter space dominated by kinetic limits on condensation. Collaborative work with Barahona and Nenes led to a refinement of the FN scheme, referred to as FN2, which now accounts for the condensational sink provided by inertially-limited giant CCN. Results from the box-model simulations demonstrate that failing to account for the condensational sink provided by the surface area of such aerosols leads to a significant overestimate of the maximum supersaturation achieved in a cloud, and hence an overestimation of the number of cloud droplets.

FN2 considerably improves the comparison with a full numerical cloud parcel model, compared to the original FN scheme. This collaborative work led to the publication of Barahona et al. (2010). The timescale of these developments was such that the choice of parameterisation for use in the climate model simulations in the rest of this thesis had to be made before the development and evaluation of FN2 was complete. Consequently, the Abdul-Razzak and Ghan (2000) was chosen for implementation in HadGEM-UKCA.

Chapter 3

The third chapter described the implementation of the Abdul-Razzak and Ghan (2000) aerosol activation scheme within HadGEM-UKCA. The ability of HadGEM-UKCA to generate realistic CCN was evaluated against measurements. 90% of modelled CCN concentrations were found to be within a factor of ten of the co-located measured values, with a tendency for the modelled values to be biased low. This appears to be a particular problem in the southeast Pacific marine stratocumulus region but the reasons for this are not certain.

Modelled aerosol optical depth (AOD), cloud droplet number concentration (CDNC), liquid cloud droplet effective radius (r_e) and cloud liquid water path (LWP) were evaluated against three Earth-observing satellite datasets. Qualitative comparisons of these quantities were made between cloud-top satellite-sampled output from HadGEM-UKCA, and similar quantities retrieved in the satellite datasets. Differences in the absolute values between the three satellite datasets can be substantial, and therefore provide little constraint on the absolute values that the model should be simulating. However, the large-scale features are similar in the three datasets, and these are mainly well replicated within the model.

Aerosol–cloud relationships within the model were quantified by calculating the sensitivities of each of the cloud properties examined—CDNC, r_e and LWP—to the AOD, following the linear regression grid method of Grandey and Stier (2010). Sensitivities and correlation coefficients were compared with those calculated within each of the three satellite datasets (Grandey, 2011). Overall, aerosol–cloud relationships consistent with both indirect aerosol effects are represented by HadGEM-UKCA. Comparison with the aerosol–cloud relationships observed in the satellite data indicate that the modelled relationships may potentially be too strong, although the low correlation coefficients of all of the relationships examined across the model and satellites mean that further evidence would be needed to quantify this.

Chapter 4

The fourth chapter examined the representation of sub-grid-scale vertical velocity within a GCM. Two distinct methods were examined: first, the use of a probability density function (pdf) to represent the variability of vertical velocity within a grid-box; and second, the use of a single characteristic updraught, representative of the typical vertical velocities associated with large-scale cloud formation within each grid-box.

A Gaussian form of pdf was selected as the baseline configuration for HadGEM-UKCA, primarily because such distributions are commonly observed in stratocumulus cloud. However, a lack of observational constraint impedes choice of a suitable function for use in a GCM and also makes it extremely difficult to evaluate any parameterisation globally. The added complexity of more elaborate functions, including modified Gaussian distributions with skewness or kurtosis incorporated, are not justifiable in a GCM until a more comprehensive and systematic set of observations is available. For these reasons, only the effect of changing the variance of the pdf (σ_w^2) was investigated further.

Five configurations of vertical velocity were defined to investigate the effects of (a) the choice of σ_w defining a pdf and (b) using a characteristic updraught instead of a pdf; three configurations were defined with differing fixed values of σ_w (spanning the observed range), one in which σ_w was derived from the modelled turbulent kinetic energy, and one with a fixed value of w .

Annual mean cloud droplet number concentration (CDNC) was found to increase (sub-linearly) with increasing σ_w , particularly in regions of high CCN concentrations. This happens because increasing the σ_w increases the weighting of higher updraught velocities. Depending on the number and size distribution of the aerosols, this tends to increase the maximum supersaturation, allowing more, smaller aerosols to activate. The effect saturates once all potential CCN have activated, and thus is stronger in regions of high CCN than low CCN. (Due to the inverse relationship with CDNC, the opposite effect was shown for liquid cloud droplet effective radius.)

Increasing σ_w was also shown to increase the liquid water path (column-integrated cloud liquid water content), due to the effects on precipitation efficiency of increased aerosol activation and hence more, smaller cloud droplets.

In the fourth vertical velocity configuration (TKE_0.1), σ_w was derived from the turbulent kinetic energy in the model, where this was greater than a threshold value. A comparison of the CDNC predicted by this configuration compared to the reference case (sigw0.4, with $\sigma_w = 0.4 \text{ m s}^{-1}$) showed that although the value of σ_w in the TKE_0.1 simulation exceeded 0.4 m s^{-1} in several regions, this did not necessarily correlate with the CDNC being higher, because such regions often occurred in areas with low CCN concentration such that aerosol activation was already saturated at $\sigma_w = 0.4 \text{ m s}^{-1}$. The most noticeable differences in CDNC between TKE_0.1 and sigw0.4 occurred in regions of moderate to high CCN concentrations, where σ_w in TKE_0.1 was less than 0.4 m s^{-1} , such that fewer aerosols could activate. One of the reasons that the TKE_0.1 underestimates CDNC compared to sigw0.4, is due to the relatively high occurrence of the minimum value of σ_w .

Model simulations with the final configuration of vertical velocity showed that using a single fixed updraught velocity of $w = 0.32 \text{ m s}^{-1}$ tends to overestimate CDNC, consistent with the findings of Morales and Nenes (2010). This study proposed that it might be adequate to determine a characteristic updraught as $w^* = \lambda \sigma_w$, but simulations with the box-model and GCM performed for this chapter demonstrated that the parameter λ is in fact highly variable, depending on the number, size distribution and composition of aerosols. Moreover, this relationship would still require σ_w to be determined at every

grid-point, which is one of the most challenging aspects of the pdf-based approach. Results from a sensitivity study showed that discretising a Gaussian pdf of updraught velocity into just 20 bins gives values of CDNC to within 1% of the value obtained with 10,000 bins, for four typical aerosol distributions and covering a representative range of values σ_w . Thus, it seems likely that the modest computational cost of using a pdf is justified.

Chapter 5

In the fifth chapter, modelled σ_w and CDNC from each of the five configurations of HadGEM-UKCA defined in Chapter 4 were compared with *in situ* measurements from flight campaigns through warm clouds at a variety of locations. As expected, the configuration in which σ_w was defined as the median of all the measurements of σ_w across these flight campaigns performed well on average in comparison to the low and high values. Another configuration in which σ_w was derived from the modelled turbulent kinetic energy (TKE) also behaved well, but the small sample size of campaigns does not provide a strong enough constraint to recommend one or the other as being most representative globally. This has implications for the best estimate of the RFP in Chapter 6, and it is likely that the uncertainty relating to this choice could be reduced by better constraint of updraught velocity through additional measurements.

Deficiencies of the TKE-dependent configuration were shown by examining high resolution time series of vertical profiles of modelled TKE, cloud-base height, CCN concentrations and resulting CDNC at each flight campaign location. However, most of these deficiencies and potential remedies are model-specific, other than the possible suggestion that the value of σ_w be regime-dependent, which may also be applicable to other models.

Chapter 6

In this chapter, radiative flux perturbations were used to quantify the radiative effects of anthropogenic aerosols in each model configuration. All of the configurations of HadGEM-UKCA investigated in this thesis predict an overall negative RFP due to anthropogenic aerosols, with estimates of the total global mean RFP ranging from -1.4 to -2.3 W m^{-2} . The majority of this range (-1.4 to -2.0 W m^{-2}) is due to varying the standard deviation of the pdf of updraught velocity from $\sigma_w = 0.1 \text{ m s}^{-1}$ to 0.7 m s^{-1} (spanning a representative range of observed values), demonstrating how great an impact this one parameter can have on the overall RFP.

The best estimate is somewhere between -1.7 and -1.9 W m^{-2} , derived from runs with a physically-realistic variable σ_w based on model turbulent kinetic energy, and a globally-fixed, mid-range value, respectively. The lack of observational constraints from the flight campaigns studied in Chapter 5 does not allow an informed choice to be made between these two estimates.

Regional variation of radiative flux perturbation was also investigated, with particular attention given to an unexpected area of positive forcing in the south-eastern Pacific. Examining this uncovered an underlying microphysical explanation, where the presence of additional Aitken- and accumulation-mode particles in a simulation with present-day aerosol emissions provides a condensational sink for atmospheric sulphuric acid vapour which inhibits nucleation of new sulphate particles.

The large uncertainty attributable to changing the distribution of updraughts, in combination with the counter-intuitive results arising from aerosol microphysics which affect

the subsequent growth of particles to CCN-range, suggests that accurate incorporation of microphysical processes is key to understanding the changing global climate.

7.2 General conclusions and recommendations for future work

Estimation of the indirect aerosol effects with a GCM is fundamentally dependent on the parameterisation of aerosol activation. Early work in this thesis highlighted the importance of having a parameterisation that fully accounts for the kinetic limitations on droplet formations. While there are surely further improvements that could be made to such parameterisations, comparisons with full numerical simulations indicate that aerosol activation is now a fairly well-parameterised process, and is not the greatest source of uncertainty in forcing estimates.

The results that can be obtained with such a parameterisation are, of course, critically dependent on the input parameters, in particular on the parameterisation of updraught velocity and the modelled aerosol distribution. Results of the simulations in Chapter 6 have highlighted the importance of two-moment aerosol microphysics schemes, since processes affecting number, mass and composition of aerosols affect their ability to act as CCN, and the interactions between such processes can be non-linear.

Analysis of data from a dozen flight campaigns, combined with a review of the literature, has demonstrated that in-cloud vertical velocity distributions in warm clouds can have a high degree of variability (even when disregarding the highly skewed distributions associated with deep convection). This high degree of variability, the large number of poorly-understood influencing factors and the sparse set of measurements makes it particularly difficult to parameterise such pdfs for GCM studies.

Simulations with HadGEM-UKCA, coupled to a Köhler-theory based aerosol activation scheme, have shown that the choice of parameterisation of sub-grid-scale vertical velocity can have a considerable impact on the predicted RFP due to indirect aerosol effects. (Using a single characteristic updraught velocity in place of a pdf is to be discouraged in GCMs, for the reasons outlined previously). In particular, the work in this thesis focused on the effect of changing the variance of a Gaussian pdf of vertical velocity (used as an input to the aerosol activation scheme within the model).

Applying a fixed value of σ_w universally is not physically realistic, but it does allow the sensitivity of the model to this parameter to be tested. Simulations with HadGEM-UKCA have shown that increasing σ_w increases the magnitude of the RFP, but that the effect is sub-linear. The RFP due to the total anthropogenic aerosol effects is estimated at -1.4 W m^{-2} from a pair of simulations with $\sigma_w = 0.1 \text{ m s}^{-1}$, up to -2.0 W m^{-2} for $\sigma_w = 0.7 \text{ m s}^{-1}$, where appropriate low and high values of σ_w have been obtained from an assessment of the range of values measured by the flight campaigns listed in Chapter 5. This range of 0.6 W m^{-2} corresponds to almost a third of the total estimate of -1.9 W m^{-2} , obtained with the mid-range value of $\sigma_w = 0.4 \text{ m s}^{-1}$.

Model simulations without the indirect aerosol effects provide an estimate of the direct effect of aerosols in this configuration of HadGEM-UKCA as -0.6 W m^{-2} . To the extent that it is legitimate to decouple the direct and indirect effects, simply subtracting this direct effect from the estimates of the total aerosol effect indicates that the range of uncertainty due to choice of σ_w is equal to nearly half (46%) of the best estimate of the RFP due to indirect effects alone (-1.3 W m^{-2}). Reducing this uncertainty in the choice of σ_w (or rather, how it is parameterised) is therefore an important step towards reducing the uncertainty in estimates of the indirect aerosol effects.

Measurements indicate that σ_w is actually highly variable between different types of clouds, and even between clouds of the same type, depending on the dominant factors influencing the updraughts (such as fluctuations in the radiative heating of the surface driving adiabatic convection, contributions from radiative cooling at the cloud-top to the in-cloud turbulence, changes to the boundary layer dynamics e.g. due to the diurnal cycle, and other sources of atmospheric turbulence, e.g. due to the effects of orography or surface roughness). Thus applying the same value of σ_w everywhere would lead to overestimates in some cases and underestimates in others. For this reason it is important to allow σ_w to be variable; preferably it should be related to other processes incorporated in the GCM that are representative of the physical drivers of this variability in the atmosphere, though a case could also be made for some kind of stochastic choice of σ_w based on an empirically (or otherwise) determined distribution of likely values, which would then need to be linked to the prognostic variables.

In one configuration tested here, σ_w was derived from the modelled turbulent kinetic energy, where this was greater than a threshold value. This led to values of σ_w that were a better match to the observations in some regions. However, the high frequency of occurrence of the minimum value meant that σ_w derived in this way tended to underpredict that observed in other cases. Overall, this leads to a total RFP of -1.7 W m^{-2} , compared to -1.9 W m^{-2} , obtained with the mid-range value of $\sigma_w = 0.4 \text{ m s}^{-1}$. Given the limited number and sparse distribution of measurements of σ_w and CDNC, there is not enough evidence to declare either of these as the best estimate of the RFP. On the one hand, it is unphysical to assume a value of $\sigma_w = 0.4 \text{ m s}^{-1}$ universally, and therefore the TKE-dependence is preferable in that it provides a more realistic representation of the spatial and temporal variability of σ_w , but on the other hand the high frequency of occurrence of the minimum value in this configuration means that it may be frequently underestimating the value of σ_w and hence possibly also the RFP.

Future plans to address this problem are two-fold. Firstly, to work on improving the parameterisation within HadGEM-UKCA by addressing some of the model-specific issues that cause the minimum value of σ_w to be hit so often (e.g. TKE is not diagnosed above the boundary layer; the definition of the lowest cloudy level). Secondly, a more comprehensive database of *in situ* measurements that systematically characterises the vertical velocity distributions in different cloud regimes is required to evaluate the parameterised values of σ_w and hence CDNC in a greater variety of conditions.

In a more general sense, the uncertainty in the indirect aerosol effects could be reduced further by the continued development of better, and potentially non-Gaussian, parameterisations of the pdf for GCM purposes. This could be derived from process studies in high-resolution numerical models, combined with more measurements. The representation of more complex distributions that capture the skewness and other higher-order moments of the distribution could also then be warranted, if the results were validated against a more comprehensive survey of in-cloud velocity distributions provided by future flight campaigns.

However, CDNC is not only affected by the updraught velocity, and some regions are more sensitive to this parameter than others. For instance, in a region of low CCN concentration with a high fraction of activation at low supersaturation, CDNC is dominated by the CCN concentration, and realistic aerosol modelling is essential for accurate predictions. This means that, should w (or σ_w) be overestimated in such a region, it would not be able to cause the predicted CDNC to be overestimated. However, in a region of high CCN concentration, the CDNC is very sensitive to the value of w (or σ_w). A metric could be devised to map out the sensitivities to each parameter within the model, potentially

by calculating $dN_{\text{act}}/d\sigma_w$. Identifying and quantifying this sensitivity by region or cloud regime is pertinent because it could be used to inform the location and focus of future flight campaigns.

In summary, predictions of the indirect radiative effects of anthropogenic aerosols on the Earth's climate are dependent on the ability of models to represent several highly non-linear microphysical processes relating to both aerosols and clouds, and the interactions between these and with the climate system at large. One of the primary aims and great challenges in climate science is to better understand, quantify and reduce the uncertainties associated with these aerosol–cloud interactions. The development of better parameterisations evaluated against more comprehensive empirical evidence, feeding into high-resolution model simulations, all have a role to play in achieving this aim.

Glossary

β	Cloud droplet spectral dispersion
ϵ_{ij}	Soluble mass fraction of component j , mode i
η	Dimensionless constant used in Abdul-Razzak and Ghan (1998, 2000) parameterisation
ν	Degree of ionic dissociation of a solute
ϕ_{ij}	Osmotic coefficient of component j , mode i
ρ_w	Density of water
σ	Surface tension
$\sigma_{g,i}$	Geometric standard deviation of aerosol mode i
σ_w	Standard deviation of vertical velocity pdf
τ_c	Cloud optical depth
ζ	Dimensionless constant used in Abdul-Razzak and Ghan (1998, 2000) parameterisation
A	Köhler Kelvin coefficient of curvature
$a_{\min}$	Radius of the smallest aerosol that activates
$a_{m,i}$	Geometric mean dry radius of aerosol mode i
B	Köhler hygroscopicity coefficient
b	Sensitivity of a cloud property to aerosol optical depth
D	Diameter
d_m	Dry count median diameter of a single log-normal aerosol mode
D_p	Equilibrium diameter of droplet at saturation
e	Vapour pressure
e_s	Saturated vapour pressure
f_c	Liquid cloud fraction
G	Gibbs free energy

- k Boltzmann constant
- L Latent heat of vaporisation
- M_w Molar mass of water
- N_{act} Fraction of activation, $N_{\text{act}} = N_a / \sum_{i=1}^I N_i$
- N_a Total number of activated aerosols
- N_d Cloud droplet number concentration
- N_i Total number concentration of aerosol in mode i
- q_c Cloud liquid water content
- r Droplet radius
- r_c Critical radius required for aerosol activation
- r_e Liquid cloud droplet effective radius
- R_v Ideal gas constant
- S Supersaturation $S = s - 1$
- s Saturation ratio
- S_c Critical supersaturation required for aerosol activation
- S_{max} Maximum supersaturation
- $S_{m,i}$ Critical supersaturation of a particle with the geometric mean radius of aerosol mode i
- T_d Dew point temperature
- w Vertical velocity
- w^* Characteristic updraught velocity
- x_{ij} Mass mixing ratio of component j , mode i
- z_{BL} Boundary layer depth
- z_{CB} Cloud-base height
- z_{TM} Maximum height of turbulent mixing
- AATSR Advanced Along-Track Scanning Radiometer
- ACE-1 The first Aerosol Characterisation Experiment (flight campaign)

- ACE-2 The second Aerosol Characterisation Experiment (flight campaign)
- AeroCOM Aerosol Comparisons between Observations and Models
- AERONET Aerosol Robotic Network
- AOD Aerosol Optical Depth
- ARG Abdul-Razzak and Ghan (2000) parameterisation
- CCN Cloud Condensation Nuclei
- CDNC Cloud Droplet Number Concentration
- CERES Clouds and the Earth's Radiant Energy System
- CLOPAP Cloud Processing of Regional Air Pollution Over Land and Sea (flight campaign)
- CMIP5 Coupled Model Intercomparison Project Phase 5
- CRYSTAL-FACE Cirrus Regional Study of Tropical Anvils and cirrus Layers–Florida Area Cirrus Experiment (flight campaign)
- ECMWF European Centre for Medium-Range Weather Forecasts
- FN Fountoukis and Nenes (2005) parameterisation
- FN2 Updated version of the FN parameterisation by Barahona, West et al. (2010)
- GCM General Circulation Model
- GlobAEROSOL ESA Data User Element project which produced a global dataset of aerosol properties from an ensemble of European satellite instruments (AATSR, AATSR-2, MERIS and SEVIRI)
- GLOMAP Global Model of Aerosol Processes
- HadGEM3-A UK Met Office Hadley Centre Global Environmental Model in atmosphere-only mode
- ICARTT The 2004 International Consortium for Atmospheric Research on Transport and Transformation experiment (flight campaign)
- IPCC Intergovernmental Panel on Climate Change
- LES Large eddy simulation
- LWP Liquid Water Path
- MODIS Moderate Resolution Imaging Spectroradiometer
- NARE North Atlantic Regional Experiment (flight campaign)
- NEPac-MASE-CSTRIPE Combination of the MARine Stratus/Stratocumulus Experiment and the Coastal STRatocumulus Imposed Perturbation Experiment (flight campaigns)
- NS Nenes and Seinfeld (2003) parameterisation

ORAC Oxford-Rutherford Appellton Laboratory Aerosol and Cloud retrieval

PASE PACific Sulfate Experiment (flight campaign)

PD Present day

PI Pre-industrial

RFP Radiative Flux Perturbation

RICO Rain in Cumulus over Ocean (flight campaign)

SOCEX Southern Ocean Cloud EXperiment (flight campaign)

SSF Single Scanner Footprint

TKE Turbulent kinetic energy

UKCA United Kingdom Chemistry and Aerosols community model

VOCALS-REx VAMOS (Variability of the American MOnsoon Systems) Ocean-Cloud-
Atmosphere-Land Study Regional Experiment (flight campaign)

Köhler theory derivation

A.1 The effect of droplet curvature

For droplets to grow, the rate of condensation of water onto the droplet must exceed the evaporation from the surface. First, consider the process of homogeneous nucleation on a spherical droplet of water. The vapour pressure over a curved surface as a function of radius, $e_s(r)$ will be discussed with respect to the vapour pressure over a plane surface, $e_s(\infty)$. Consider the change in Gibbs free energy due to the formation of a single droplet of radius r containing n molecules of water

$$\Delta G = G_{\text{droplet}} - G_{\text{pure vapour}}. \quad (\text{A.1})$$

Let the total initial number of molecules be N_T , and the number of molecules remaining in the vapour state after the droplet forms be $N_T - n = N_v$. Let g_v and g_l be the Gibbs free energies of a molecule in the vapour or liquid state, respectively. Then

$$\Delta G = N_v g_v + n g_l + 4\pi r^2 \sigma - N_T g_v, \quad (\text{A.2})$$

where $4\pi r^2 \sigma$ represents the work required to create the spherical surface between the liquid and vapour, and σ is the surface tension. This may be rewritten as

$$\Delta G = n(g_l - g_v) + 4\pi r^2 \sigma. \quad (\text{A.3})$$

The number of molecules in the droplet is $n = \frac{4}{3}\pi r^3 / v_l$, where v_l is the volume occupied by a molecule in the liquid phase. This can be included to give

$$\Delta G = \frac{4\pi r^3}{3v_l} (g_l - g_v) + 4\pi r^2 \sigma. \quad (\text{A.4})$$

The Gibbs free energy G is defined as

$$G = U + pV + TS \quad (\text{A.5})$$

and chemical potential, μ_i of a species i is defined by

$$\mu_i = \left(\frac{\partial U}{\partial n_i} \right)_{S, V, n_j} \quad (\text{A.6})$$

where n_i is the number of moles of species i . By using the differential form of equation A.5 and combining with the differential form of the thermodynamic relation for the internal energy of a system of variable composition, incorporating the chemical potential from equation A.6 we obtain

$$dG = -SdT + Vdp + \sum_{i=1}^k \mu_i dn_i. \quad (\text{A.7})$$

At constant temperature, and fixed number of molecules, this reduces to $dg = vdp$ or $dg = (v_l - v_v)dp$. In the relevant limit $v_v \gg v_l$, so neglecting v_l relative to v_v gives $dg = -v_v dp$. Assume the vapour to be an ideal gas so $v_v = kT/p$. Integrating gives

$$g_l - g_v = -kT \int_{e_s(\infty)}^e \frac{dp}{p} = -kT \ln \left(\frac{e}{e_s(\infty)} \right). \quad (\text{A.8})$$

Defining the saturation ratio, $s = e/e_s(\infty)$ and substituting equation A.8 into equation A.4 gives the following expression for the Gibbs free energy associated with homogeneous nucleation of a spherical droplet

$$\Delta G = -\frac{4\pi r^3}{3v_l} kT \ln s + 4\pi r^2 \sigma. \quad (\text{A.9})$$

The sign of the first term on the right hand side is defined by the saturation ratio, and this has a strong effect on the behaviour of ΔG as a function of r . In sub-saturated conditions, $e < e_s(\infty)$ so $\ln s$ is negative, and hence ΔG is always positive and increases with increasing r . This inhibits droplet formation under sub saturated conditions. In supersaturated conditions, $e > e_s(\infty)$ so $\ln s$ is positive. In this case, ΔG can be positive or negative, depending on the value of r . Initially, ΔG increases with increasing r , inhibiting the growth of very small particles in supersaturated conditions. ΔG reaches a maximum at $r = r_c$ and then decreases with further increase in r , allowing a droplet to continue growing spontaneously through condensation from the vapour phase, since further growth causes a further drop in ΔG . The maximum is a metastable equilibrium, and its location can be found by setting $\partial \Delta G / \partial r = 0$ in equation A.9 to obtain

$$r_c = \frac{2\sigma v_l}{kT \ln s}. \quad (\text{A.10})$$

This equation relates the equilibrium radius of a droplet of pure substance (in our case, water) to the physical properties of that substance, σ and v_l , and the saturation ratio over the droplet in its environment. It can be re-expressed in terms of molar units as the more familiar Kelvin equation

$$\frac{e_s(r)}{e_s(\infty)} = \exp \left(\frac{2\sigma M_w}{\rho_w R T r} \right) = \exp \left(\frac{A}{r} \right), \quad (\text{A.11})$$

where

$$A = \frac{2\sigma M_w}{\rho_w R T}. \quad (\text{A.12})$$

It is apparent from the Kelvin equation that the vapour pressure over a curved surface of a substance is always greater than the vapour pressure over a plane surface of the same substance (Seinfeld and Pandis, 1998; Wallace and Hobbs, 2006).

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