Cloud cycling and scavenging of aerosols in the Unified Model and UKCA-MODE

Zak Kipling

August 2010

Supervised by Philip Stier¹ and Colin Johnson²

D. Phil. first year report
~ 10500 words

¹Atmospheric, Oceanic and Planetary Physics, University of Oxford, UK
²Met Office Hadley Centre, Exeter, UK
## Contents

<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>Contents</td>
<td>2</td>
</tr>
<tr>
<td>List of Tables</td>
<td>4</td>
</tr>
<tr>
<td>List of Figures</td>
<td>5</td>
</tr>
<tr>
<td><strong>1 Introduction</strong></td>
<td>6</td>
</tr>
<tr>
<td>1.1 Aerosols in the climate system</td>
<td>6</td>
</tr>
<tr>
<td>1.1.1 Nature, sources and geographical distribution</td>
<td>6</td>
</tr>
<tr>
<td>1.1.2 Direct radiative effect</td>
<td>8</td>
</tr>
<tr>
<td>1.1.3 Indirect effects via cloud and precipitation</td>
<td>8</td>
</tr>
<tr>
<td>1.2 Removal processes: scavenging and deposition</td>
<td>10</td>
</tr>
<tr>
<td>1.2.1 Nucleation scavenging</td>
<td>10</td>
</tr>
<tr>
<td>1.2.2 Impaction scavenging</td>
<td>11</td>
</tr>
<tr>
<td>1.2.3 Previous experiments with detailed scavenging schemes</td>
<td>11</td>
</tr>
<tr>
<td>1.3 Parameterisation of aerosol, cloud and precipitation in GCMs</td>
<td>11</td>
</tr>
<tr>
<td>1.3.1 Size distribution</td>
<td>11</td>
</tr>
<tr>
<td>1.3.2 Mixing state</td>
<td>12</td>
</tr>
<tr>
<td>1.4 Cloud cycling</td>
<td>13</td>
</tr>
<tr>
<td>1.5 Cloud, precipitation and aerosols in the Unified Model</td>
<td>14</td>
</tr>
<tr>
<td>1.5.1 Cloud and precipitation</td>
<td>14</td>
</tr>
<tr>
<td>1.5.2 Aerosols: UKCA-MODE</td>
<td>14</td>
</tr>
<tr>
<td>1.6 Aims of this project</td>
<td>14</td>
</tr>
<tr>
<td><strong>2 Cloud cycling in the Unified Model and UKCA-MODE</strong></td>
<td>16</td>
</tr>
<tr>
<td>2.1 Diagnosing the cloud water cycle</td>
<td>16</td>
</tr>
<tr>
<td>2.2 Diagnosing cycling of aerosol through clouds</td>
<td>18</td>
</tr>
<tr>
<td>2.3 Results</td>
<td>20</td>
</tr>
<tr>
<td><strong>3 Developing a microphysically-consistent scavenging scheme for UKCA</strong></td>
<td>27</td>
</tr>
<tr>
<td>3.1 Problems with the current scheme</td>
<td>27</td>
</tr>
<tr>
<td>3.2 An improved, microphysically consistent scheme</td>
<td>28</td>
</tr>
<tr>
<td>3.3 Mathematical formulation</td>
<td>31</td>
</tr>
<tr>
<td>3.3.1 Processes releasing aerosol from water phases to free/interstitial modes</td>
<td>34</td>
</tr>
<tr>
<td>3.3.2 Processes taking up free/interstitial aerosol to water phases</td>
<td>40</td>
</tr>
<tr>
<td>3.3.3 Implementation in modal aerosol scheme</td>
<td>41</td>
</tr>
<tr>
<td>3.4 Discretisation</td>
<td>42</td>
</tr>
</tbody>
</table>
## List of Tables

1.1 Categorisation of scavenging processes ............................................ 10

2.1 Comparison of global annual cloud water and in-cloud aerosol budgets as simulated by the Unified Model with UKCA-MODE with estimates derived from ECHAM5-HAM and global budget arguments ........................................... 25

2.2 As Table 2.1 for UM/UKCA, for each aerosol component. ......................... 26

3.1 Differences between the chemistry and aerosol in-cloud scavenging schemes in UKCA. .......................................................... 29

3.2 Transfer processes in the large-scale precipitation scheme in the Unified Model 30

3.3 Overview of a consistent in-cloud wet deposition scheme for UKCA. ............ 32
List of Figures

1.1 Global mean radiative forcings from different mechanisms, with estimated uncertainties. ........................................ 7
1.2 The atmospheric sulphur cycle leading to the production of sulphate (SO$_4^{2-}$) aerosol. ........................................ 8
2.1 Global-mean large-scale cloud water budget for one year, as simulated by the Unified Model. ................................ 21
2.2 Global-mean budgets for in-cloud (left) and total (right) aerosol mass and number for one year, as simulated by the Unified Model with UKCA-MODE. . . . . . . 22
2.3 Time evolution of global aerosol mass and number (left) and mean burden over one year (right), as simulated by the Unified Model with UKCA-MODE. . . . . . 23
Chapter 1

Introduction

1.1 Aerosols in the climate system

Aerosol particles (small solid or liquid particles suspended in the atmosphere) play an important role in both global and regional climate, both through absorption and scattering of radiation (Section 1.1.2) and interaction with clouds and precipitation (Section 1.1.3). Aerosol particles come from a variety of natural and anthropogenic sources, vary substantially in size and chemical composition, and have a very non-uniform geographical distribution (Section 1.1.1).

Aerosol effects are some of the largest uncertainties in estimates of total anthropogenic radiative forcing (Forster et al., 2007), as shown in Figure 1.1; they are thought to be a negative forcing, but the extent to which they presently cancel the positive forcing from greenhouse gases is poorly quantified. There are also many complex interactions and feedbacks between aerosol processes and other elements of the climate system, which have only recently begun to be represented in climate modelling studies (Carslaw et al., 2010). It is thus important to develop a better understanding of aerosol processes, and their effects on climate, if we are to reduce these uncertainties.

1.1.1 Nature, sources and geographical distribution

Aerosol particles are described as either primary (emitted in particulate form at source) or secondary (produced from atmospheric trace gases by nucleation/condensation and/or chemical reactions). Although fresh primary aerosol may have a very specific composition determined by its source, particles are typically of mixed composition due to coagulation between particles and condensation of one substance onto the surface of another (Andreae et al., 2008).

The size spectrum of atmospheric aerosol ranges from freshly-nucleated (mostly sulphate) particles a few nm in diameter up to coarse mechanically-produced particles many µm in diameter. In between lie what are termed Aitken particles (10 nm–100 nm), and accumulation-mode particles (100 nm–1 µm). The accumulation and coarse modes account for most of the aerosol mass in the atmosphere; however the smaller nucleation-mode and Aitken particles are much more numerous.

Sulphates

While some sulphate (SO$_4^{2-}$) aerosol may be emitted directly in particulate form, the majority is formed from atmospheric SO$_2$ either by oxidation by OH to H$_2$SO$_4$ followed by nucleation/condensation, or by oxidation in the aqueous phase (within cloud droplets) by O$_3$ or H$_2$O$_2$ to produce dissolved aerosol (Seinfeld and Pandis, 2006, §22.1). SO$_2$ is itself produced by oxidation of a variety of precursor gases – in particular hydrogen sulphide (H$_2$S), dimethyl
1.1. AEROSOLS IN THE CLIMATE SYSTEM

Figure 1.1: Global mean radiative forcings from different mechanisms, with estimated uncertainties. Taken from Forster et al. (2007, FAQ 2.1, fig. 2).

sulphide (DMS, CH\textsubscript{3}SCH\textsubscript{3}), carbon disulphide (CS\textsubscript{2}) and carbonyl sulphide (OCS) – as well as by direct emission (Seinfeld and Pandis, 2006, §2.2). Figure 1.2 shows a representation of the atmospheric sulphur cycle. DMS (produced in the oceans by phytoplankton) accounts for much of the natural sulphate precursor emissions, while the (larger) anthropogenic contribution comes from the burning of fossil fuel (especially coal) in the form of SO\textsubscript{2}.

**Sea salt**

Over the ocean, salt particles released by the evaporation of sea spray are a significant source of aerosol particles, composed largely of sodium chloride (NaCl); the rate at which such particles are produced is strongly dependent on surface wind speed.

**Carbon**

A wide variety of carbon compounds are found in aerosol particles; however, these are usually divided into two broad classes:

- **black carbon** (or elemental carbon), strongly light-absorbing graphite-like material produced during combustion as a principal component of soot; and

- **organic carbon**, consisting of biological debris and secondary aerosol from chemical processing and condensation of organic gases of both natural and anthropogenic origin.
CHAPTER 1. INTRODUCTION

Figure 1.2: The atmospheric sulphur cycle leading to the production of sulphate ($\text{SO}_4^{2-}$) aerosol. Based on Seinfeld and Pandis (2006, fig. 22.2).

Mineral dust

Mineral dust is carried into the air by wind blowing over soil or sand; over and downwind of arid regions in particular, this contributes a large proportion of the aerosol population.

Nitrogen compounds

Both ammonia ($\text{NH}_3$) and nitric acid ($\text{HNO}_3$) vapour may condense onto aerosol particles, dissociating to form ammonium ($\text{NH}_4^+$) and nitrate ($\text{NO}_3^-$) ions (Andreae et al., 2008, §3.2.6). Although they can form a significant fraction of total aerosol mass in some regions (Jimenez, 2009, fig. 1), and both their potential effect on the nitrogen cycle and their contribution to aerosol direct and indirect effects has been recognised (Denman et al., 2007, §7.4.2), these aerosol components have received relatively little attention in climate modelling studies.

1.1.2 Direct radiative effect

Aerosol particles can directly alter the radiative balance of the atmosphere by absorbing or scattering incoming solar radiation. Scattering aerosols effectively increase the planetary albedo, leading to a cooling effect. Absorbing aerosols (e.g. black carbon, as the name implies) may have a cooling or warming effect, depending on the albedo of the underlying surface. The net direct radiative effect of anthropogenic aerosol is estimated to be a cooling one, although its magnitude is rather uncertain (Schulz et al., 2006; Forster et al., 2007).

1.1.3 Indirect effects via cloud and precipitation

Aerosol particles can also alter the climate through their roles in cloud and precipitation microphysics:
1.1. AEROSOLS IN THE CLIMATE SYSTEM

- cloud condensation nuclei (CCN) are required for new liquid cloud droplets to form from water vapour (Rogers and Yau, 1989, ch. 6);
- ice nuclei (IN) are required for new ice particles to form directly from water vapour, or by the freezing of supercooled liquid cloud droplets at temperatures above about \(-40^\circ\text{C}\) (Rogers and Yau, 1989, ch. 9).

Changes in these processes can alter both the radiative properties and precipitation efficiency of clouds; together these are termed indirect effects and are reviewed in Lohmann and Feichter (2005) and Denman et al. (2007, §7.5.2).

Two main mechanisms for such an indirect effect have been identified and studied: the cloud albedo effect and the cloud lifetime effect. A number of other mechanisms, mostly involving mixed-phase cloud, have also been proposed but have not been studied in such detail.

### Cloud albedo effect

The size distribution of cloud droplets is heavily dependent on the number of CCN available; an increase in the number of suitable aerosol particles will lead to a greater number of smaller droplets for a given liquid water content. In most cases this leads to an increase in cloud albedo (Twomey, 1977), thus having an overall cooling effect.

### Cloud lifetime effect

As well as increasing the cloud albedo, the reduction in droplet size is likely to delay the onset of precipitation in shallow low-level cloud, which requires the development of much larger drops (Albrecht, 1989). This will increase the lifetime of such clouds; assuming that the availability of water vapour is not a limiting factor (which is unlikely at least in maritime environments), this in turn increases the mean coverage of low-level cloud. Such cloud has little effect on outgoing long-wave radiation, but has a high short-wave albedo, thus exerting a further cooling effect.

### Other indirect effects

There are a number of other proposed mechanisms for indirect effects, including:

- The “glaciation indirect effect” (Lohmann, 2002), where an increase in the number of aerosol particles acting as ice nuclei enhances glaciation in mixed-phase clouds, thus increasing precipitation and reducing their lifetime.
- The “de-activation effect”, where increased sulphate forms a coating on particles that would otherwise act as ice nuclei, preventing them from doing so (Pruppacher and Klett, 1996, §9.2).
- The “riming indirect effect”, where smaller cloud droplets lead to less riming of ice in mixed-phase clouds (Borys et al., 2003).
- Thermodynamic effects: smaller cloud droplets freeze at lower temperatures, delaying the onset of glaciation and precipitation in mixed-phase clouds (Rosenfeld and Woodley, 2000; Khain et al., 2001); consequent changes in latent heat release may invigorate or reduce further convection (Khain et al., 2005).

These additional effects involving mixed-phase clouds are somewhat less well understood than the cloud albedo and cloud lifetime effects; however at least one recent study (Lohmann and Hoose, 2009) suggests that these may contribute significantly to the total anthropogenic aerosol indirect effect.
CHAPTER 1. INTRODUCTION

Table 1.1: Categorisation of scavenging processes

<table>
<thead>
<tr>
<th></th>
<th>Nucleation scavenging</th>
<th>Impaction scavenging</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>In-cloud</strong></td>
<td>Activation as cloud condensation or ice nuclei</td>
<td>Collision with cloud droplets or ice crystals</td>
</tr>
<tr>
<td><strong>Below-cloud</strong></td>
<td>—</td>
<td>Collision with falling precipitation</td>
</tr>
</tbody>
</table>

There are also so-called “semi-direct” effects, where cloud properties and atmospheric stability are influenced by the local atmospheric heating caused by absorbing aerosol particles; these are reviewed in Koch and Del Genio (2010).

With the exception of the cloud albedo effect, none of the indirect or semi-direct effects can be properly considered radiative forcings in the usual sense as they do not cause instantaneous changes to the radiation budget, but involve feedbacks via the hydrological cycle; this, along with the high level of uncertainty in these effects, is why they do not appear in the IPCC Fourth Assessment Report’s summary of radiative forcings (Figure 1.1). In order to quantify the full range of aerosol indirect effects, we need to use an alternative methodology such as radiative flux perturbation (Haywood et al., 2009; Lohmann et al., 2010) which can take into account feedback processes.

In addition, it is important to note that many anthropogenic trace gases and aerosols are known or suspected to be harmful to human health, and therefore emission controls are likely to be desirable irrespective of their effects on climate.

1.2 Removal processes: scavenging and deposition

There are two main routes by which aerosol particles are removed from the atmosphere, referred to as dry and wet deposition. In dry deposition, aerosol particles settle out due to gravity, diffusion and other processes and eventually attach to the surface. This process is not the subject of this study, but is described in more detail in e.g. Ruijgrok et al. (1995). In wet deposition, aerosol particles are taken up (“scavenged”) by hydrometeors and carried to the surface in precipitation.

There are a variety of scavenging processes, which are usually divided either into in-cloud scavenging (by cloud droplets or ice crystals) and below-cloud scavenging (by precipitation), or into nucleation scavenging (where aerosol particles act as cloud condensation or ice nuclei) and impaction scavenging (where aerosol particles collide with existing hydrometeors). Nucleation scavenging only occurs in-cloud, while impaction scavenging occurs both in- and below-cloud (see Table 1.1).

Note that scavenged aerosol is not necessarily removed from the atmosphere by wet deposition at the surface: if cloud droplets evaporate (or ice crystals sublime) without forming precipitation, or if precipitation evaporates/sublimes before reaching the surface, then the aerosol is released back to the atmosphere. This cloud/precipitation processing of aerosols is discussed in more detail in Section 1.4.

1.2.1 Nucleation scavenging

Nucleation scavenging occurs when aerosol particles form cloud condensation or ice nuclei, and thus become dissolved or embedded in the resulting hydrometeor.
For liquid cloud this process, described by Köhler theory, depends on the aerosol composition and size distribution and local supersaturation (Rogers and Yau, 1989, ch. 6; Pruppacher and Klett, 1996, §9.1). The theory is well established, and detailed parameterisations have been developed to represent the activation process in models with size-resolved aerosol (e.g. Nenes and Seinfeld, 2003; Fountoukis and Nenes, 2005), as well as more parameterised schemes (e.g. Abdul-Razzak and Ghan, 2000).

Heterogeneous nucleation of ice is somewhat more complex, with the crystal structure of the aerosol particles (which may depend on both the composition and history of the particle) also playing an important role (Pruppacher and Klett, 1996, §9.2; Cantrell and Heymsfield, 2005). A theoretical parameterisation thus requires knowledge of details of the aerosol microphysical state which are not usually available in current models; thus current modelling generally relies on empirical relationships such as those of DeMott et al. (2010).

### 1.2.2 Impaction scavenging

Impaction scavenging occurs when aerosol particles collide and coalesce with hydrometeors, thus becoming dissolved, attached or embedded. This can be driven by a number of processes, which dominate in different regions of the joint aerosol-hydrometeor size spectrum, including Brownian diffusion, interception, inertial impaction, electrostatic and phoretic effects (Pruppacher and Klett, 1996, §17.4.2).

An assessment of the uncertainties in current parameterisations of impaction scavenging by raindrops by these processes is given by Wang (2010); they conclude that estimates of the collision efficiency between droplets and aerosol particles in the 0.01 µm to 3 µm range are the major contributor, with theoretical parameterisations an order of magnitude or more below that required to match observed scavenging rates. Increased scavenging due to turbulent flow (Grover and Pruppacher, 1985; Pinsky and Khain, 1997; Khain and Pinsky, 1997) is suggested as a possible cause of this mismatch between theoretical parameterisations (usually assuming non-turbulent flow) and observations (usually subject to boundary-layer turbulence).

Parameterisations of impaction scavenging by ice and snow are less well developed, and tend to be heavily dependent on the shape of the ice particles. A detailed discussion of the theory and available observations can be found in Wang (2002, ch. 5), and Feng (2009) presents a physically-based parameterisation using up-to-date estimates of the relevant parameters.

### 1.2.3 Previous experiments with detailed scavenging schemes

Croft et al. (2009, 2010b) consider the effect of introducing a detailed size-resolved scheme for impaction scavenging by rain and cloud droplets in the ECHAM5-HAM model, along with a highly parameterised activation scheme for nucleation scavenging, and show significant improvements in simulated aerosol distribution compared to a simple scheme using prescribed scavenging ratios for in-cloud scavenging and a first-order loss rate for impaction scavenging.

### 1.3 Parameterisation of aerosol, cloud and precipitation in General Circulation Models

#### 1.3.1 Size distribution

Many microphysical processes involving aerosol particles and/or hydrometeors are heavily dependent on their size distribution, and not just on their total mass mixing ratio in the atmosphere. For example:
CHAPTER 1. INTRODUCTION

- surface processes (e.g. growth by condensation or deposition) depend more on total surface area (which is greater for smaller particles at a given total mass);
- there is a critical size at which aerosol particles can activate to form cloud droplets, dependent on their composition and the local supersaturation of water vapour according to Köhler theory;
- large cloud droplets are more likely to grow to form raindrops;
- smaller raindrops are more likely to evaporate before reaching the surface.

In simple “bulk” schemes, only the total mass of each type of aerosol particle or hydrometeor is transported, and either a fixed or diagnosed size distribution is assumed when parameterising the microphysical processes; this assumed distribution may be tuned individually for each process to match observations without necessarily having a physical justification. While rain may be distinguished from cloud, or coarse aerosol from fine, there is no way to capture changes of particle size within these broad categories.

Increasing computing power has made it possible to represent the size distribution prognostically, allowing it to evolve according to the microphysical and transport processes rather than assuming a distribution at each timestep based on diagnostic relationships. There are two main approaches to this: bin-resolved schemes and multi-moment modal schemes.

In bin-resolved schemes, the size spectrum for each particle type is divided into a large number of discrete “bins” each covering a relatively narrow size range; the number or mass of particles in each bin is then transported separately, with explicit representation of the microphysical processes that grow or shrink particles from one bin to another. While such schemes are able to represent the evolution of size distributions very well, the large number of prognostic variables required makes them very expensive in terms of computing time and storage when used in global models.

Multi-moment modal schemes attempt to capture the evolving size distribution with a much smaller number of variables than required for a bin scheme. The size distribution of each type of particle is assumed to be a superposition of a small number of “modes” with prescribed (usually gamma or log-normal) distributions. The moments of the distribution for each mode are then carried as prognostic variables:

- with one moment, the distribution is fixed, and only the strength of each mode can change – this is essentially the same as the bulk schemes mentioned above;
- with two moments, typically number (zeroth moment) and mass (proportional to the third moment), the mean particle size in each mode can vary, but the distribution shape still remains fixed;
- with additional moments, the width and shape of each mode can also vary.

Microphysical processes are then represented in terms of integrals over the distribution for each mode, to calculate their effects on these moments. A mode-merging algorithm may be used to transfer particles from the tail of one mode into an adjacent one if their overlap becomes too great.

1.3.2 Mixing state

As mentioned in Section 1.1.1, aerosol particles are generally of mixed composition. This has important implications for both their radiative and microphysical properties (Seinfeld and Pandis, 2006, §24.6; Levin and Brenguier, 2008, §6.2.1); parameterisations which assume
separate populations of sulphate and black carbon aerosol ("externally-mixed"), for example, may behave quite differently to those which assume they are well mixed in the same particles ("internally-mixed").

There is also the matter of aerosol/water mixing: not only do hydrometeors generally contain some dissolved or embedded aerosol material, but hygroscopic aerosol particles (e.g. sulphate) usually contain some condensed water even if they are too small to activate as cloud droplets. Some recent studies (e.g. Ovchinnikov and Easter, 2010) have used a two-dimensional joint distribution to capture the evolution of the aerosol/water mixing state; while such an approach can be useful in a limited-area model, the increased computational cost makes it unsuitable for use in a global model.

There is also a distinction to be made between models which transport in-droplet and in-ice aerosol as separate prognostics (and thus remember the activation history) and those which diagnose the in-cloud fraction afresh at each timestep. Hoose et al. (2008a,b) show that introducing a prognostic treatment to ECHAM5-HAM has a significant impact on simulated aerosol distributions. Ghan and Easter (2006) examine a range of approaches from fully prognostic to fully diagnostic in-cloud aerosol, using an otherwise identical model setup. They show little difference between the fully-prognostic and the non-advected configurations, but significant local error and global bias when using diagnostic in-cloud aerosol.

1.4 Cloud cycling

A significant proportion of cloud water is not removed by precipitation, but returned to the atmosphere as water vapour by evaporation; Pruppacher and Jaenicke (1995) estimate (based on a global budget argument) that cloud water goes through 10 condensation-evaporation cycles on average before falling to the ground as precipitation. Since the formation of cloud droplets (or ice crystals) involves the take-up of aerosol particles as CCN or IN (and further particles may be taken up by impaction scavenging), it follows that some of this material will be released from evaporating clouds and precipitation. However, the physical and chemical characteristics of these released particles may be different to those of the scavenged particles: cloud droplets typically undergo many collision/coalescence events during their lifetime, thus each evaporating droplet will in general release an aerosol particle containing the material from several CCN and/or IN, plus that taken up from impaction scavenging or produced by aqueous chemistry. Thus cloud processing can be expected to cause a shift from small aerosols with diverse composition towards larger aerosols with a well-mixed composition.

Pruppacher and Jaenicke (1995) estimate that a typical aerosol particle in the atmosphere has been cycled through cloud three times, thus suggesting that cloud processing is likely to play a significant role in the aerosol life cycle.

Hoose et al. (2008a) diagnose the cloud cycling rate for stratiform clouds only using the ECHAM5-HAM model. Despite significant differences in aerosol emissions and liquid water path from the values used by Pruppacher and Jaenicke (1995), the stratiform cloud-cycling rates are in fair agreement.

It is worth noting, however, that the approach of Pruppacher and Jaenicke (1995) suggests that most of the cloud cycling occurs in convective cloud – the rates are reduced from 10 to 2.6 times for water and from 3 to 0.4 times for aerosol particles when only stratiform cloud is considered. Croft et al. (2010a) have extended the work of Hoose et al. (2008a) to include cycling through convective cloud; their results suggest a more equal split between cycling in stratiform and convective clouds.
1.5 Cloud, precipitation and aerosols in the Unified Model

1.5.1 Cloud and precipitation

Recent versions of the UK Met Office Unified Model (UM) implement a prognostic cloud scheme (PC2, Wilson et al., 2008, 2010) for large-scale (as opposed to sub-grid-scale convective) cloud. This treats liquid water content, ice content and (liquid/ice/mixed) cloud fraction as prognostic variables allowing clouds to evolve and be transported between timesteps. As a one-moment bulk scheme, there is no tracking of the cloud droplet size distribution; the detailed microphysics of the large-scale precipitation scheme (Wilson and Ballard, 1999; Wilkinson et al., 2009) uses assumed size distributions based on diagnostic relationships.

Sub-grid-scale convection is parameterised by a mass-flux scheme (Gregory and Rowntree, 1990; Gregory et al., 2009) which diagnoses both convective precipitation and vertical transport of tracers and momentum. In PC2-based configurations, convective cloud cover (e.g. for radiation) is treated by detraining cloud water into the large-scale cloud scheme, rather than using a separate convective cloud fraction.

1.5.2 Aerosols: UKCA-MODE

Older versions of the UM (e.g. HadGEM2) implement an externally-mixed bulk aerosol scheme (Jones et al., 2001; Johnson et al., 2007; Bellouin et al., 2007). Recent versions also contain the newer UKCA-MODE scheme (Mann et al., 2010), which uses an internally-mixed two-moment (mass and number) modal representation.

In the current standard configuration, four aerosol components (sulphate, sea salt, black carbon and organic carbon) are tracked in four soluble modes (nucleation, Aitken, accumulation and coarse) and one insoluble mode (Aitken). There are also options for a mineral dust component (which is still in development, and adds insoluble accumulation and coarse modes) and separate tracking of primary and secondary organic carbon. Nitrate and ammonium are not currently included.

Transport of aerosol by large-scale dynamics, convection and boundary-layer mixing is handled by the generic tracer transport scheme in the UM. Explicit parameterisations are added for aerosol microphysics (nucleation, condensation, coagulation and ageing), removal processes (dry and wet deposition) and in-cloud chemical production of secondary aerosol. The process parameterisations are derived from those used in the bin-resolved GLOMAP scheme (Spracklen et al., 2005).

While UKCA-MODE offers a number of improvements compared to the old scheme, the implementation of in-cloud scavenging is quite simplistic. This will be discussed in more detail in Chapter 3, but in particular there is no prognostic representation of dissolved aerosol, no scavenging by snow, and no re-release of aerosol when rain evaporates. Below-cloud scavenging by rain is more detailed, using a lookup table of collection efficiencies for different raindrop and aerosol particle sizes; however once again neither scavenging by snow nor evaporation of rain is considered.

1.6 Aims of this project

The estimates of Pruppacher and Jaenicke (1995) suggest that cloud processing plays a significant role in the life cycle of atmospheric aerosol. As an initial experiment, in Chapter 2 we attempt to quantify this in the context of a general circulation model (GCM) with a coupled aerosol scheme, specifically UKCA-MODE running within the Met Office Unified Model.
For many aerosol components, wet deposition in precipitation is the dominant removal process; thus its efficiency has a strong effect on their lifetime in the atmosphere and how far from the source regions they are transported. It has been shown (e.g. Bourgeois and Bey, 2010) that the aerosol distribution in regions far from sources is very sensitive to small changes in scavenging parameters in a highly-parameterised scheme. One of the questions we hope to answer is how the long-range transport under a physically-detailed scavenging scheme compares with that achieved by empirical tuning of a more parameterised scheme – in particular, can we simulate realistic transport while reducing the degree of tuning required? In order to investigate this, a more detailed scavenging scheme will be needed; this is discussed in Chapter 3.

We also hope to investigate the impact of scavenging on the vertical transport of aerosol – in particular, removal by convective precipitation may cause a significant reduction in the effective vertical transport of aerosol by convective plumes, and scavenging followed by re-evaporation represents a downward transport of aerosol. Vertically-resolved observations, e.g. from aircraft campaigns, will be crucial for evaluating modelled vertical distributions in this context.

Finally, cloud cycling of aerosols is likely to have an impact on both the direct and indirect aerosol effects, which we aim to quantify once we have developed a detailed model of the cycling process. As mentioned earlier, the aerosol particles released on evaporation are likely to be larger than those originally scavenged due to coalescence between hydrometeors. This shift in size distribution will alter both the radiative properties of the aerosol (modifying the direct effect), and the number of CCN or IN available for subsequent cloud formation (modifying the indirect effect).
Chapter 2

Cloud cycling in the Unified Model and UKCA-MODE

In this chapter, we develop a method for diagnosing the rates at which both water and aerosol are cycled through large-scale cloud (as described in Section 1.4) in the Unified Model (UM) and UKCA-MODE, presenting results from a one-year simulation which are then compared with those from previous studies.

2.1 Diagnosing the cloud water cycle

Following Pruppacher and Jaenicke (1995), the number of re-evaporation cycles is given by the ratio of evaporation of cloud water to precipitation reaching the surface. The latter is easily obtained from standard model output; for large-scale cloud the former can be calculated by separating out the water vapour increments due to phase changes in the atmosphere (as opposed to transport or surface fluxes). Because convective cloud only exists transiently within the model, there are no readily-available diagnostics for the evaporation of sub-grid-scale convective cloud; thus only cycling through large-scale cloud is considered in this analysis.

In the PC2 cloud scheme, prognostic variables for (grid-box mean) vapour ($q$), liquid ($q_{cl}$) and ice ($q_{cf}$) evolve through increments from each of the dynamical and physical processes in the model (Wilson et al., 2008, §4):

$$
\frac{\partial q_x}{\partial t} = \frac{\partial q_x}{\partial t}_{\text{adv}} + \frac{\partial q_x}{\partial t}_{\text{rad}} + \frac{\partial q_x}{\partial t}_{\text{conv}} + \frac{\partial q_x}{\partial t}_{\text{exp}} + \frac{\partial q_x}{\partial t}_{\text{bl}} + \frac{\partial q_x}{\partial t}_{\text{chk}}
$$

for $q_x \in \{q, q_{cl}, q_{cf}\}$.

The terms in red (advection by the model dynamics) do not correspond to any evaporation or condensation of cloud, only its transport from one grid box to another (and thus conserve globally integrated $q_x$). These can be ignored when calculating the condensation/evaporation rate.

Those in blue (radiation, adiabatic expansion due to large scale ascent, cloud initiation and bounds checking\(^1\)) represent pure phase change processes (and thus conserve total grid-box water $q + q_{cl} + q_{cf}$). Thus each process $P$ contributes

$$
(E - C)|_P = \frac{\partial q}{\partial t}|_P - \frac{\partial q_{cl}}{\partial t}|_P - \frac{\partial q_{cf}}{\partial t}|_P
$$

\(^1\)As described in Wilson et al. (2010, §4.10), small adjustments may be made to the $q_x$ and $C_x$ prognostics to ensure physical consistency, for example by evaporating any liquid water where $C_1 = 0$. 

16
2.1. DIAGNOSING THE CLOUD WATER CYCLE

to the net (evaporation − condensation) rate. (Sublimation and deposition of ice cloud are thus included, as if they occurred via the liquid phase.)

The terms in green, however, represent more complex schemes (convection, large-scale precipitation and the boundary layer) which include both phase change and transport or surface exchange processes:

- Convective cloud water/ice is considered transient in the model; it does not appear in $q_{cl}$ or $q_{cf}$, except for that which is detrained from the convective plumes as large-scale cloud. From a large-scale perspective, there is a vertical transport of water vapour (and possibly cloud water or ice if they are entrained) in the convective plumes, a sink of this water to the surface in convective precipitation, a downward transport due to subsidence to balance the upward mass flux in the plumes, and condensation where water or ice is detrained:

$$\frac{\partial q_x}{\partial t} \bigg|_{\text{conv}} = \frac{\partial q_x}{\partial t} \bigg|_{\text{conv plume}} + \frac{\partial q_x}{\partial t} \bigg|_{\text{conv precip}} + \frac{\partial q_x}{\partial t} \bigg|_{\text{conv subsidence}} + \frac{\partial q_x}{\partial t} \bigg|_{\text{conv detrain}}. \quad (2.3)$$

Because the individual contributions of these processes are not easily separable in the model diagnostics, we assume that the contributions from entrainment and subsidence of large-scale water and ice are small, and thus

$$(E - C)\bigg|_{\text{conv}} \approx -\frac{\partial q_{cl}}{\partial t} \bigg|_{\text{conv}} - \frac{\partial q_{cf}}{\partial t} \bigg|_{\text{conv}}. \quad (2.4)$$

This approximation is not ideal, and could certainly be improved on in the context of a more detailed approach to convective diagnostics which would also allow analysis of condensation-evaporation cycling in convective cloud.

- The large-scale precipitation scheme includes increments to $q_x$ due to falling/settling of water and ice (i.e. downward transport of $q_{cl}$ and $q_{cf}$, but not $q$), in-cloud freezing/melting (exchange between $q_{cl}$ and $q_{cf}$), removal of water and ice as precipitation (loss of $q_{cl}$ and $q_{cf}$), ice deposition (conversion of $q$ to $q_{cf}$) and evaporation/sublimation (conversion of $q_{cl}$, $q_{cf}$ or rain to $q$). Thus, although $q_{cl}$ and $q_{cf}$ are subject to transport and surface exchange processes, the only increments to $q$ are due to evaporation and deposition/sublimation of ice:

$$\frac{\partial q}{\partial t} \bigg|_{\text{lsp}} = \frac{\partial q}{\partial t} \bigg|_{\text{lsp evap cloud}} + \frac{\partial q}{\partial t} \bigg|_{\text{lsp evap rain}} + \frac{\partial q}{\partial t} \bigg|_{\text{lsp dep ice}} + \frac{\partial q}{\partial t} \bigg|_{\text{lsp sub ice}}, \quad (2.5)$$

and thus we can calculate the contribution as

$$(E - C)\bigg|_{\text{lsp}} = \frac{\partial q}{\partial t} \bigg|_{\text{lsp}}. \quad (2.6)$$

- The boundary layer scheme includes increments from surface exchange (principally evaporation of surface water), turbulent mixing (transport of $q_x$) and condensation resulting from adiabatic expansion during this mixing:

$$\frac{\partial q}{\partial t} \bigg|_{\text{bl}} = \frac{\partial q}{\partial t} \bigg|_{\text{bl surf}} + \frac{\partial q}{\partial t} \bigg|_{\text{bl mix}} + \frac{\partial q}{\partial t} \bigg|_{\text{bl cond}}. \quad (2.7)$$

Additional diagnostics have been added to isolate this third term, so that the boundary layer contribution can be calculated as

$$(E - C)\bigg|_{\text{bl}} = \frac{\partial q}{\partial t} \bigg|_{\text{bl cond}} = -\frac{\partial q_{cl}}{\partial t} \bigg|_{\text{bl cond}} - \frac{\partial q_{cf}}{\partial t} \bigg|_{\text{bl cond}}. \quad (2.8)$$
Thus the net (evaporation − condensation) rate of large-scale cloud in each grid box can be calculated as:\(^2\):

\[
(E - C) \approx \frac{\partial q}{\partial t}_{\text{rad}} - \frac{\partial q_{\text{cl}}}{\partial t}_{\text{conv}} + \frac{\partial q_{\text{cf}}}{\partial t}_{\text{conv}} + \frac{\partial q}{\partial t}_{\text{lsp}} + \frac{\partial q}{\partial t}_{\text{bl}} + \frac{\partial q}{\partial t}_{\text{exp}} + \frac{\partial q}{\partial t}_{\text{chk}}.
\]

In grid boxes where this is positive, we diagnose evaporation; where it is negative, condensation. It is tempting to split the contribution from each process separately into positive (evaporation) and negative (condensation) components in an attempt to capture at least some cases where both occur in the same grid box. However, doing so appears misguided since a warming process (e.g., absorption of solar radiation) might actually inhibit the condensation due to a cooling process (e.g., adiabatic expansion), rather than evaporating the water after it condenses.

### 2.2 Diagnosing cycling of aerosol through clouds

It is somewhat harder to diagnose the rate at which aerosol particles in UKCA-MODE are cycled through cloud, since there is no prognostic treatment of the split between in-cloud and interstitial aerosol; thus there are no increments which can be analysed as for the water cycle. Instead, we begin by defining this split diagnostically following the approach of the in-cloud nucleation scavenging parameterisation: a fixed scavenging ratio \(R_{\text{scav}, M}\) is defined for each mode \(M\), so that the in-cloud fraction of both number and mass in that mode is given by \(CR_{\text{scav}, M}\) where \(C\) is the grid-box cloud fraction; the in-cloud mass of aerosol species \(i\) is then

\[
m_{c}^{(i)} = \sum_{\mathcal{M} \in \mathcal{M}} CR_{\text{scav}, M} m_{M}^{(i)}, \tag{2.10}
\]

where \(\mathcal{M}\) is the set of aerosol modes and \(m_{M}^{(i)}\) is the mass of aerosol species \(i\) in mode \(M\). In standard configurations,

\[
R_{\text{scav}, M} = \begin{cases} 
1 & \text{for the soluble accumulation and coarse modes} \\
0 & \text{for all other modes},
\end{cases} \tag{2.11}
\]

i.e., in the presence of cloud all aerosol in these two modes is considered to be taken up into the cloud water, while all aerosol in other modes is considered to be interstitial.

Differentiating, we can calculate the change of this diagnosed in-cloud aerosol mass as

\[
\frac{\partial m_{c}^{(i)}}{\partial t} = \sum_{\mathcal{M} \in \mathcal{M}} R_{\text{scav}, M} \left( m_{M}^{(i)} \frac{\partial C}{\partial t} + C \frac{\partial m_{M}^{(i)}}{\partial t} \right), \tag{2.12}
\]

splitting it into changes due to changing cloud cover and aerosol microphysics. (Because the diagnosed in-cloud aerosol fraction depends on cloud fraction but not cloud water content, there is no change due to condensation or evaporation unless the cloud fraction changes; this would not be the case for a fully prognostic treatment of in-cloud aerosol.) The change in

\[^{2}\text{This worked fine in UM version 7.1; in 7.3 the lowest model level shows a very large source of} \ q \text{ due to short-wave radiation. This appears to be a bug in the model diagnostics, and should not be taken to represent evaporation of cloud water; by (2.2) we can replace} \ \frac{\partial q}{\partial t}_{\text{rad}} \text{ with} \ - \frac{\partial q_{\text{cl}}}{\partial t}_{\text{rad}} - \frac{\partial q_{\text{cf}}}{\partial t}_{\text{rad}}, \text{ which appears to exclude this effect.} \]
cloud cover can be diagnosed similarly to the evaporation/condensation rate, by taking the non-transport increments to $C$ from PC2:

$$\frac{\partial C}{\partial t} \bigg|_{\text{non-transport}} \approx \frac{\partial C}{\partial t} \bigg|_{\text{rad}} + \frac{\partial C}{\partial t} \bigg|_{\text{conv}} + \frac{\partial C}{\partial t} \bigg|_{\text{lsp}}$$

$$+ \frac{\partial C}{\partial t} \bigg|_{\text{bl cond}} + \frac{\partial C}{\partial t} \bigg|_{\text{exp}} + \frac{\partial C}{\partial t} \bigg|_{\text{init}} + \frac{\partial C}{\partial t} \bigg|_{\text{chk}}$$

(again assuming the transport component of the convective increment is small). Increasing cloud cover implies take-up of aerosol in the newly-cloudy fraction of the grid-box, while decreasing cloud cover implies release in the newly-clear fraction. The changes due to aerosol microphysics can be split according to the individual processes:

$$\frac{\partial m^{(i)}_M}{\partial t} \bigg|_{\text{non-transport}} = \frac{\partial m^{(i)}_M}{\partial t} \bigg|_{\text{prim}} + \frac{\partial m^{(i)}_M}{\partial t} \bigg|_{\text{nucl}} + \frac{\partial m^{(i)}_M}{\partial t} \bigg|_{\text{cond}} + \frac{\partial m^{(i)}_M}{\partial t} \bigg|_{\text{wetox}}$$

$$+ \frac{\partial m^{(i)}_M}{\partial t} \bigg|_{\text{nuc scav}} + \frac{\partial m^{(i)}_M}{\partial t} \bigg|_{\text{imp scav}} + \frac{\partial m^{(i)}_M}{\partial t} \bigg|_{\text{dry dep}}$$

$$+ \frac{\partial m^{(i)}_M}{\partial t} \bigg|_{\text{coag}} + \frac{\partial m^{(i)}_M}{\partial t} \bigg|_{\text{age}} + \frac{\partial m^{(i)}_M}{\partial t} \bigg|_{\text{merge}};$$

however the contribution of these processes to aerosol take-up/release needs considering carefully.

With the exception of wet (in-cloud) oxidation, the source processes (primary emissions, nucleation and condensation) are straightforward: a fraction $CR_{\text{scav},M}$ of the aerosol produced in each mode $M$ is assumed to be taken up into the cloud. Wet oxidation, however, can only take place in-cloud; thus the full amount produced is assumed to be taken up, but a fraction $(1 - CR_{\text{scav},M})$ immediately released.

Aerosol removed by dry deposition is assumed to be interstitial; thus the fraction $CR_{\text{scav},M}$ implicitly removed from the diagnostic in-cloud aerosol mass is assumed to be released immediately prior to removal. (In fact, the dry deposition term also includes a vertical transport component due to gravitational settling; this is not currently diagnosed separately and, like the transport component of the convective $q_x$ and $C$ increments, assumed to be small. It should, however, be treated by considering the change in scavenged fraction across the grid-box boundary.

Nucleation scavenging, like wet oxidation, can only take place in-cloud; thus the fraction $(1 - CR_{\text{scav},M})$ implicitly removed from the diagnostic interstitial aerosol is assumed to be taken up immediately prior to removal. (This is particularly important where convective scavenging occurs but there is little large-scale cloud.) Impaction scavenging may remove either interstitial or in-cloud aerosol; no take-up or release is inferred, but separate diagnostics are introduced to identify the contribution to the in-cloud aerosol budget.

The inter-modal processes (coagulation, ageing and mode-merging) are assumed to take up or release aerosol if the scavenging ratios for the source and destination modes differ: for a transfer from $M'$ to $M$,

$$\frac{\partial m^{(i)}_M}{\partial t} \bigg|_{M' \rightarrow M} = (R_{\text{scav},M} - R_{\text{scav},M'}) \frac{\partial m^{(i)}_M}{\partial t} \bigg|_{M' \rightarrow M}. \quad (2.15)$$
Putting all these terms together, we can derive the total rates of aerosol take-up into, and release from, cloud water as

\[ T^{(i)} = \sum_{M \in M} \left\{ R_{\text{scav}, M} m_{M}^{(i)} \left[ \frac{\partial C}{\partial t} \right]_{\text{non-transport}} + CR_{\text{scav}, M} \left( \frac{\partial m_{M}^{(i)}}{\partial t} \right)_{\text{prim}} + \frac{\partial m_{M}^{(i)}}{\partial t} \left|_{\text{nuc}} \right. + \frac{\partial m_{M}^{(i)}}{\partial t} \left|_{\text{cond}} \right) \right. \]

\[ + \frac{\partial m_{M}^{(i)}}{\partial t} \left|_{\text{wetox}} \right. + (1 - CR_{\text{scav}, M}) \frac{\partial m_{M}^{(i)}}{\partial t} \left|_{\text{scav}} \left) \right. \right. \]

\[ + \sum_{M' \in M_{M}^{(i)} < R_{\text{scav}, M}} \frac{R_{\text{scav}, M} - R_{\text{scav}, M'}}{R_{\text{scav}, M}} \times \left( \frac{\partial m_{M}^{(i)}}{\partial t} \left|_{\text{coag}} \right. \right. \]

\[ + \frac{\partial m_{M}^{(i)}}{\partial t} \left|_{\text{age}} \right. + \frac{\partial m_{M}^{(i)}}{\partial t} \left|_{\text{merge}} \right) \right. \right. \}

\( (2.16) \)

and

\[ R^{(i)} = - \sum_{M \in M} \left\{ R_{\text{scav}, M} m_{M}^{(i)} \left[ \frac{\partial C}{\partial t} \right]_{\text{non-transport}} + CR_{\text{scav}, M} \frac{\partial m_{M}^{(i)}}{\partial t} \right. \]

\[ + (1 - CR_{\text{scav}, M}) \frac{\partial m_{M}^{(i)}}{\partial t} \left|_{\text{wetox}} \right. + CR_{\text{scav}, M} \frac{\partial m_{M}^{(i)}}{\partial t} \left|_{\text{dry dep}} \right) \right. \]

\[ + \sum_{M' \in M_{M}^{(i)} < R_{\text{scav}, M}} \frac{R_{\text{scav}, M} - R_{\text{scav}, M'}}{R_{\text{scav}, M}} \times \left( \frac{\partial m_{M}^{(i)}}{\partial t} \left|_{\text{coag}} \right. \right. \]

\[ + \frac{\partial m_{M}^{(i)}}{\partial t} \left|_{\text{age}} \right. + \frac{\partial m_{M}^{(i)}}{\partial t} \left|_{\text{merge}} \right) \right. \}

\( (2.17) \)

such that the in-cloud aerosol budget is

\[ \frac{\partial m_{c}^{(i)}}{\partial t} \left|_{\text{non-transport}} = T^{(i)} - R^{(i)} + \frac{\partial m_{c}^{(i)}}{\partial t} \left|_{\text{scav}} + \frac{\partial m_{c}^{(i)}}{\partial t} \left|_{\text{imp}} \right) \right. \right. \]

\( (2.18) \)

2.3 Results

The results presented here are taken from a proto-HadGEM3-A base configuration at UM version 7.3, running at N96L38 resolution (1.25° latitude × 1.875° longitude × 38 vertical levels) with the UKCA-MODE aerosol scheme added, plus additional diagnostic code required for the analysis described above. The model was spun up for three months from 1 September 1999, with results taken over the following 12 months (December 1999 – November 2000 inclusive).

Figure 2.1 shows the global-mean large-scale cloud water budget over the year, calculated as described above. From this, it appears that approximately as much large-scale cloud water is lost to evaporation as to precipitation.

Figure 2.2 shows the budgets for in-cloud and total aerosol mass and number. From the total-aerosol budgets, it appears that (large-scale) in-cloud nucleation scavenging dominates the removal of sulphate (SU) and organic carbon (OC) mass, and to a lesser extent black
carbon (BC), while dry deposition is the dominant removal process for sea salt (SS). The removal of aerosol particle number is split almost evenly between these two processes, while below-cloud impaction scavenging and convective nucleation scavenging make a much smaller contribution. The in-cloud budgets show that more in-cloud aerosol is released by evaporating cloud than is removed by scavenging processes, with the possible exception of sea salt where the split is almost even.

There are small residuals in both the diagnosed cloud water and aerosol budgets, which do not always match the net change over the year; these may in part be due to the approximate treatment of some processes, as discussed in the preceding sections. However, they are in general small compared to the dominant processes in the water and aerosol cycle, so any impact on our results should be minor.

Figure 2.3 shows the time evolution and geographical distribution of the aerosol burden. Two anomalous spikes in the sea salt burden are evident, off the west coast of South America and the southeast coast of Greenland. These are unrealistic, and thought to be caused by problems with operator-splitting between dry deposition and boundary-layer mixing (Colin Johnson, 2010, personal communication). Given the rapid high-frequency changes in the sea-salt burden compared to the other species, it is unsurprising that it should be more susceptible to such operator-splitting artifacts.

Table 2.1 compares these results to those of Hoose et al. (2008a), obtained using the ECHAM5-HAM model with a prognostic in-cloud aerosol scheme (their “AP” simulation), as well as the simple global budget estimates of Pruppacher and Jaenicke (1995), from where the notation used originates. While the estimate of cloud water lifetime with respect to precipitation ($\tau_{c,p} = \text{TWP}/P = 2.0 \text{h}$) is in the same range as the other estimates, the cloud water cycling rate ($E_{c,c}/P = 1.01$ times) is about half (and the lifetime with respect to evaporation...
Figure 2.2: Global-mean budgets for in-cloud (left) and total (right) aerosol mass and number for one year, as simulated by the Unified Model with UKCA-MODE. (SU = sulphate; SS = sea salt; BC = black carbon; OC = organic carbon.)
Figure 2.3: Time evolution of global aerosol mass and number (left) and mean burden over one year (right), as simulated by the Unified Model with UKCA-MODE. The “residual” figures are the slopes of straight lines (plotted in green) from initial to final value, which should match the time-averaged budget residual over the same period. (SU = sulphate; SS = sea salt; BC = black carbon; OC = organic carbon.)
about four times) that of Hoose et al. (2008a); the cycling rate is even further from that of Pruppacher and Jaenicke (1995). The aerosol cloud cycling rate \( E_{\text{AP,c}} / S_{\text{AP}} = 0.15 \) is also much lower – less than half that of Pruppacher and Jaenicke (1995), and just over a quarter that of Hoose et al. (2008a). The total aerosol mass source is larger than that used by Hoose et al. (2008a), while the mass released by evaporation is much smaller. The former is in spite of the current omission of mineral dust from UKCA-MODE; the latter may be partially explained by the lack of release due to evaporation of rain (which probably explains the lower \( E_{\text{AP,c}} / C_{\text{AP,c}} \) and a much larger below-cloud scavenging rate, as well as reduced take-up into the cloud water.

It is worth noting that these figures vary significantly for different aerosol species, as shown in Table 2.2. In particular, it appears that the low cycling rate obtained from UKCA-MODE is dominated by the behaviour of sea-salt, which comprises over 97% of the total aerosol mass source but is mostly removed quickly by dry deposition so that it accounts for less than half the total mass burden. The other species show much larger cycling rates, with the estimate of Pruppacher and Jaenicke (1995) well within the spanned range.

Thus overall aerosol cloud cycling rates calculated in this way are heavily dependent on the relative emissions of the different species – a factor which is not considered in the simple analysis of Pruppacher and Jaenicke (1995) where a homogeneous aerosol population is assumed. Although Hoose et al. (2008a) use a model which treats the different aerosol species explicitly, they do not discuss their different cloud-cycling rates. In their extension to convective cycling however, Croft et al. (2010a) observe a similar variation, with sea salt (and dust, which is not included in UKCA at present) undergoing significantly fewer cloud cycles than sulphate and carbonaceous aerosol.
Table 2.1: Comparison of global annual cloud water and in-cloud aerosol budgets as simulated by the Unified Model with UKCA-MODE with estimates derived from ECHAM5-HAM (Hoose et al., 2008a) and global budget arguments (Pruppacher and Jaenicke, 1995).

<table>
<thead>
<tr>
<th></th>
<th>P&amp;J&lt;sup&gt;a&lt;/sup&gt;</th>
<th>Hoose&lt;sup&gt;b&lt;/sup&gt;</th>
<th>UM/UKCA&lt;sup&gt;c&lt;/sup&gt;</th>
</tr>
</thead>
<tbody>
<tr>
<td>TWP / g m&lt;sup&gt;-2&lt;/sup&gt;</td>
<td>136</td>
<td>64</td>
<td>105</td>
</tr>
<tr>
<td>P / g m&lt;sup&gt;-2&lt;/sup&gt; h&lt;sup&gt;-1&lt;/sup&gt;</td>
<td>57</td>
<td>55</td>
<td>52</td>
</tr>
<tr>
<td>C&lt;sub&gt;v,c&lt;/sub&gt; / g m&lt;sup&gt;-2&lt;/sup&gt; h&lt;sup&gt;-1&lt;/sup&gt;</td>
<td>206</td>
<td>155</td>
<td>101</td>
</tr>
<tr>
<td>E&lt;sub&gt;v,c&lt;/sub&gt; / g m&lt;sup&gt;-2&lt;/sup&gt; h&lt;sup&gt;-1&lt;/sup&gt;</td>
<td>148</td>
<td>100</td>
<td>52</td>
</tr>
<tr>
<td>E&lt;sub&gt;v,c&lt;/sub&gt; / C&lt;sub&gt;v,c&lt;/sub&gt;</td>
<td>0.72</td>
<td>0.64</td>
<td>0.52</td>
</tr>
<tr>
<td>E&lt;sub&gt;v,c&lt;/sub&gt; / P</td>
<td>2.6</td>
<td>1.8</td>
<td>1.01</td>
</tr>
<tr>
<td>τ&lt;sub&gt;c,p&lt;/sub&gt; / h</td>
<td>2.4</td>
<td>1.2</td>
<td>2.0</td>
</tr>
<tr>
<td>τ&lt;sub&gt;c,e&lt;/sub&gt; / min</td>
<td>55</td>
<td>38</td>
<td>121</td>
</tr>
<tr>
<td>m&lt;sub&gt;AP&lt;/sub&gt; / Tg</td>
<td>3</td>
<td>34.9&lt;sup&gt;d&lt;/sup&gt;</td>
<td>8.20</td>
</tr>
<tr>
<td>m&lt;sub&gt;AP,ic&lt;/sub&gt; / Tg</td>
<td></td>
<td>-</td>
<td>0.684</td>
</tr>
<tr>
<td>C&lt;sub&gt;AP,e&lt;/sub&gt; / Tg yr&lt;sup&gt;-1&lt;/sup&gt;</td>
<td>1072</td>
<td>5596</td>
<td>3279</td>
</tr>
<tr>
<td>E&lt;sub&gt;AP,e&lt;/sub&gt; / Tg yr&lt;sup&gt;-1&lt;/sup&gt;</td>
<td>775</td>
<td>4027</td>
<td>1618</td>
</tr>
<tr>
<td>W&lt;sub&gt;dep,ic&lt;/sub&gt; / Tg yr&lt;sup&gt;-1&lt;/sup&gt;</td>
<td>297</td>
<td>1570</td>
<td>1011</td>
</tr>
<tr>
<td>W&lt;sub&gt;dep,bc&lt;/sub&gt; / Tg yr&lt;sup&gt;-1&lt;/sup&gt;</td>
<td>74</td>
<td>237</td>
<td>1011</td>
</tr>
<tr>
<td>W&lt;sub&gt;dep&lt;/sub&gt; / Tg yr&lt;sup&gt;-1&lt;/sup&gt;</td>
<td>371</td>
<td>1826</td>
<td>2022</td>
</tr>
<tr>
<td>D&lt;sub&gt;dep&lt;/sub&gt; / Tg yr&lt;sup&gt;-1&lt;/sup&gt;</td>
<td>1639&lt;sup&gt;e&lt;/sup&gt;</td>
<td>5750&lt;sup&gt;e&lt;/sup&gt;</td>
<td>8972</td>
</tr>
<tr>
<td>S&lt;sub&gt;AP&lt;/sub&gt; / Tg yr&lt;sup&gt;-1&lt;/sup&gt;</td>
<td>2000</td>
<td>7576</td>
<td>10786</td>
</tr>
<tr>
<td>E&lt;sub&gt;AP,e&lt;/sub&gt; / C&lt;sub&gt;AP,e&lt;/sub&gt;</td>
<td>0.72</td>
<td>0.72</td>
<td>0.49</td>
</tr>
<tr>
<td>E&lt;sub&gt;AP,e&lt;/sub&gt; / S&lt;sub&gt;AP&lt;/sub&gt;</td>
<td>0.4</td>
<td>0.53</td>
<td>0.15</td>
</tr>
<tr>
<td>τ&lt;sub&gt;AP,p&lt;/sub&gt; / h</td>
<td>71</td>
<td>167</td>
<td>35.6</td>
</tr>
<tr>
<td>τ&lt;sub&gt;AP,e&lt;/sub&gt; / h</td>
<td>34</td>
<td>75.9</td>
<td>44.4</td>
</tr>
<tr>
<td>τ&lt;sub&gt;AP,d&lt;/sub&gt; / h</td>
<td>16</td>
<td>53.1</td>
<td>8.0</td>
</tr>
<tr>
<td>τ&lt;sub&gt;AP,ic,p&lt;/sub&gt; / h</td>
<td>-</td>
<td>-</td>
<td>5.93</td>
</tr>
<tr>
<td>τ&lt;sub&gt;AP,ic,e&lt;/sub&gt; / h</td>
<td>-</td>
<td>-</td>
<td>3.71</td>
</tr>
</tbody>
</table>

| TWP                           | total water path (liquid + ice) |
| P                             | large-scale precipitation rate at surface |
| C<sub>v,c</sub>              | condensation rate of cloud water |
| E<sub>v,c</sub>              | evaporation rate of cloud water and precipitation |
| τ<sub>c,p</sub>              | lifetime of cloud water with respect to precipitation, = TWP/P |
| τ<sub>c,e</sub>              | lifetime of cloud water with respect to evaporation, = TWP/E<sub>v,c</sub> |
| m<sub>AP</sub>               | total mass of aerosol |
| m<sub>AP,ic</sub>            | total mass of in-cloud aerosol |
| C<sub>AP,e</sub>             | take-up rate of aerosol into cloud water |
| E<sub>AP,e</sub>             | release rate of aerosol from cloud water |
| W<sub>dep,ic</sub>           | in-cloud scavenging rate of aerosol |
| W<sub>dep,bc</sub>           | below cloud scavenging rate of aerosol |
| D<sub>dep</sub>              | dry deposition rate of aerosol |
| W<sub>dep</sub>              | total scavenging rate of aerosol, = W<sub>dep,ic</sub> + W<sub>dep,bc</sub> |
| S<sub>AP</sub>               | total aerosol source (from primary emissions and secondary production) |
| τ<sub>AP,p</sub>             | lifetime of aerosol with respect to precipitation, = m<sub>AP</sub>/W<sub>dep</sub> |
| τ<sub>AP,e</sub>             | lifetime of aerosol with respect to release by evaporation, = m<sub>AP</sub>/E<sub>AP,e</sub> |
| τ<sub>AP,d</sub>             | lifetime of aerosol with respect to dry deposition, = m<sub>AP</sub>/D<sub>dep</sub> |
| τ<sub>AP,ic,p</sub>          | lifetime of in-cloud aerosol with respect to precipitation, = m<sub>AP,ic</sub>/W<sub>dep,ic</sub> |
| τ<sub>AP,ic,e</sub>          | lifetime of in-cloud aerosol with respect to release by evaporation, = m<sub>AP,ic</sub>/E<sub>AP,e</sub> |

<sup>a</sup>Hoose et al. (2008a), using ECHAM5-HAM with a prognostic in-cloud aerosol scheme, for all stratiform clouds
<sup>b</sup>Pruppacher and Jaenicke (1995), recalculated for stratiform clouds only following Hoose et al. (2008a)
<sup>c</sup>diagnosed from the UM/UKCA-MODE as described in the text, for all stratiform clouds
<sup>d</sup>treating sulphate aerosol as H<sub>2</sub>SO<sub>4</sub>, as in UKCA
<sup>e</sup>these figures are not given, but assuming a closed budget in equilibrium S<sub>AP</sub> = W<sub>dep</sub> + D<sub>dep</sub>
Table 2.2: As Table 2.1 for UM/UKCA, for each aerosol component. (SU = sulphate; SS = sea salt; BC = black carbon; OC = organic carbon.)

<table>
<thead>
<tr>
<th></th>
<th>SU</th>
<th>SS</th>
<th>BC</th>
<th>OC</th>
</tr>
</thead>
<tbody>
<tr>
<td>$m_{AP}$ / Tg</td>
<td>1.97</td>
<td>4.46</td>
<td>0.138</td>
<td>1.63</td>
</tr>
<tr>
<td>$m_{AP,ic}$ / Tg</td>
<td>0.122</td>
<td>0.501</td>
<td>0.00474</td>
<td>0.0565</td>
</tr>
<tr>
<td>$C_{AP,c}$ / Tg yr$^{-1}$</td>
<td>400</td>
<td>2624</td>
<td>20.9</td>
<td>234</td>
</tr>
<tr>
<td>$E_{AP,c}$ / Tg yr$^{-1}$</td>
<td>297</td>
<td>1148</td>
<td>12.4</td>
<td>160</td>
</tr>
<tr>
<td>$W_{dep,ic}$ / Tg yr$^{-1}$</td>
<td>107</td>
<td>842</td>
<td>5.19</td>
<td>57.1</td>
</tr>
<tr>
<td>$W_{dep,ic}$ / Tg yr$^{-1}$</td>
<td>2.70</td>
<td>1005</td>
<td>0.261</td>
<td>3.00</td>
</tr>
<tr>
<td>$W_{dep}$ / Tg yr$^{-1}$</td>
<td>109</td>
<td>1846</td>
<td>5.45</td>
<td>60.1</td>
</tr>
<tr>
<td>$D_{dep}$ / Tg yr$^{-1}$</td>
<td>12.0</td>
<td>8934</td>
<td>3.43</td>
<td>22.2</td>
</tr>
<tr>
<td>$S_{AP}$ / Tg yr$^{-1}$</td>
<td>134</td>
<td>10550</td>
<td>9.66</td>
<td>92.5</td>
</tr>
<tr>
<td>$E_{AP,c}$ / $C_{AP,c}$</td>
<td>0.74</td>
<td>0.44</td>
<td>0.59</td>
<td>0.68</td>
</tr>
<tr>
<td>$E_{AP,c}$ / $S_{AP}$</td>
<td>2.22</td>
<td>0.11</td>
<td>1.28</td>
<td>1.73</td>
</tr>
<tr>
<td>$E_{AP,c}$ / $W_{dep,ic}$</td>
<td>2.79</td>
<td>1.36</td>
<td>2.39</td>
<td>2.80</td>
</tr>
<tr>
<td>$\tau_{AP,p}$ / h</td>
<td>158</td>
<td>21.2</td>
<td>223</td>
<td>238</td>
</tr>
<tr>
<td>$\tau_{AP,e}$ / h</td>
<td>58.1</td>
<td>34.0</td>
<td>97.8</td>
<td>89.6</td>
</tr>
<tr>
<td>$\tau_{AP,d}$ / h</td>
<td>1444</td>
<td>4.37</td>
<td>353</td>
<td>644</td>
</tr>
<tr>
<td>$\tau_{AP,ic,p}$ / h</td>
<td>9.99</td>
<td>5.22</td>
<td>8.00</td>
<td>8.68</td>
</tr>
<tr>
<td>$\tau_{AP,ic,e}$ / h</td>
<td>3.58</td>
<td>3.83</td>
<td>3.35</td>
<td>3.10</td>
</tr>
</tbody>
</table>
Chapter 3

Developing a microphysically-consistent scavenging scheme for UKCA

In Chapter 1 we alluded to some shortcomings of the current scavenging scheme in UKCA-MODE; in this chapter we first discuss these in more detail and then derive and present a more physically-detailed approach, aiming for consistency with the large-scale precipitation scheme. This is motivated by a desire to investigate the extent to which such a scheme can give improved predictions of global aerosol distributions (particularly with regard to long-range transport) on a physical basis, without a heavy reliance on empirically-tuned coefficients.

3.1 Problems with the current scheme

The current aerosol scavenging scheme in UKCA is quite simplistic. In particular:

- Nucleation scavenging assumes a fixed proportion of each mode is taken up by the cloud water; this proportion is the same for mass and number (equivalent to scavenging uniformly across the size spectrum of the mode), and is not dependent on a detailed parameterisation of droplet activation.

- Nucleation scavenging assumes a fixed removal timescale in any grid-box where rain is formed (i.e. flux out > flux in); no account is taken of the cloud fraction or precipitation rate, although these are readily available from the cloud and precipitation schemes.

- Impaction scavenging by cloud droplets is ignored, although this is understood to be an important removal mechanism for particles too small to act as CCN or IN.

- Neither nucleation or impaction scavenging by snow is considered; the former incorrectly occurs where snow melts to form rain.

- Scavenged aerosol contained in evaporating precipitation is not returned to the atmosphere; this may lead to removal being too fast, and also ignores a mechanism for downward transport.

- Convective transport and scavenging are operator-split, such that aerosol can be transported above the level at which it should be scavenged and detrained before the scavenging scheme sees it; this may slow down removal and exaggerate upward transport.
The tropospheric chemistry component of UKCA (O’Connor, 2009) also implements in-cloud scavenging for trace gases; while the mechanism of take-up into cloud water is different from that for aerosols, there are several apparently arbitrary differences in the treatment of precipitation microphysics in the two schemes, summarised in Table 3.1. In a coupled chemistry-aerosol scheme, it is clearly desirable that both should be removed by precipitation in a consistent way.

3.2 An improved, microphysically consistent scheme

The goal of the scheme presented here is to treat wet deposition of chemical and aerosol species by large-scale precipitation in a consistent way, taking account of the detailed microphysical processes diagnosed by the large-scale precipitation scheme. It should also be implemented such that it can interface with the convection scheme in the future (with the addition of the necessary hooks and additional diagnostics therein) to provide consistent convective scavenging.

The PC2 large-scale cloud scheme (Wilson et al., 2010) partitions each grid box into clear, liquid, ice and mixed-phase volume fractions ($C_0$, $C_l$, $C_i$ and $C_{mp}$ respectively). The precipitation scheme (Wilkinson et al., 2009) adds a rain volume fraction $C_R$ which overlaps with these (in a well-defined order of preference: $C_l \rightarrow C_{mp} \rightarrow C_i \rightarrow C_0$).

The water in the gridbox is split into vapour, liquid cloud, ice crystals, snow aggregates, rain and graupel ($q$, $q_{cl}$, $q_{cfc}$, $q_{cfa}$, $q_R$, and $q_{grau}$ respectively). In typical climate configurations, only $q$, $q_{cl}$ and a combined $q_{cf} = q_{cfc} + q_{cfa}$ exist as prognostics, and graupel is turned off completely; however, there are options for prognostic rain, separate ice/snow prognostics and (prognostic) graupel. (This is assuming PC2 is in use; if PC2 is turned off, even $q_{cl}$ is not prognostic. The new scavenging scheme is not intended to be used in non-PC2 configurations, and they will not be considered further here.)

The large-scale precipitation scheme calculates mass transfer rates for a number of separate microphysical processes which transfer mass between these phases, as shown in Table 3.2, as well as fall rates for each phase.

The new scheme is based on the idea that, once taken up by liquid water or ice, a chemical or aerosol species should “follow” that water through these processes, until one of the following occurs:

1. The water returns to the vapour phase. In this case, a proportion of aerosol species is released corresponding to that contained within completely-evaporated droplets (or completely-sublimated ice particles). Chemical species are taken up or released by liquid water according to effective Henry’s law as a “fast” process as the liquid water content changes. Specific notes on the different vapour-phase processes are given in Table 3.2.

2. The water falls out of the grid-box, taking any contained chemical or aerosol with it into the grid-box below, or depositing it at the surface if it falls out of the lowest level.

3. The water remains in the grid-box at the end of the timestep, in which case any contained chemical or aerosol is also retained in the grid-box. (In a prognostic treatment, this would be retained separately from gas-phase chemical or interstitial aerosol, however in the present framework this distinction is lost between timesteps.)

At the start of the timestep, chemical species are taken up into liquid water according to effective Henry’s law. Aerosol is taken up into liquid water and ice according to scavenging coefficients which may be either fixed for each aerosol mode and water phase, or derived from the number of activated droplets (for nucleation scavenging).
3.2. AN IMPROVED, MICROPHYSICALLY CONSISTENT SCHEME

Table 3.1: Differences between the chemistry and aerosol in-cloud scavenging schemes in UKCA.

<table>
<thead>
<tr>
<th>Formulation of parent scheme</th>
<th>Updating</th>
<th>Chemistry</th>
<th>Aerosols</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Process rate calculated and solved in parallel with other chemical processes.</td>
<td>Process directly updates tracer amount in a sequential fashion.</td>
<td></td>
</tr>
<tr>
<td>Grid</td>
<td>Working on 3D arrays.</td>
<td>Working on 1D arrays.</td>
<td></td>
</tr>
<tr>
<td>Intrinsic differences</td>
<td>Aqueous fraction</td>
<td>Effective Henry’s law for chemical species, using fixed cloud liquid water content.</td>
<td>Prescribed scavenging coefficient for aerosol mode.</td>
</tr>
<tr>
<td>Arbitrary differences</td>
<td>Relation to precipitation rate $P_k$ out of layer</td>
<td>Empirical linear relation of removal rate to $\sum_{k'=k}^{k_{max}} P_{k'}$. (counting precipitation from higher levels multiple times).</td>
<td>Prescribed removal timescale applied where any rain is formed (i.e. $P_k &gt; P_{k+1}$).</td>
</tr>
<tr>
<td>Convection</td>
<td>Large-scale and convective precipitation handled independently.</td>
<td>Scavenging by convective precipitation only where no large-scale precipitation is formed; otherwise the latter is assumed to dominate.</td>
<td></td>
</tr>
<tr>
<td>Ice</td>
<td>Linear minimum temperature cut-off (between $0^\circ$C and $-20^\circ$C) for removal by large-scale precipitation in polar regions (beyond $65^\circ$).</td>
<td>Insoluble modes not scavenged unless $T &lt; -15.15^\circ$C (though never with default scavenging ratios).</td>
<td></td>
</tr>
<tr>
<td>Snow</td>
<td>Included in $P_k$: treated identically to rain.</td>
<td>Not included in $P_k$: treated as newly-formed rain where it melts.</td>
<td></td>
</tr>
</tbody>
</table>
Table 3.2: Transfer processes in the large-scale precipitation scheme in the Unified Model (based on Wilkinson et al., 2009, Table 11).

<table>
<thead>
<tr>
<th>Process</th>
<th>From $\rightarrow$ To $\rightarrow$ To</th>
<th>Description</th>
<th>Note</th>
</tr>
</thead>
<tbody>
<tr>
<td>LSETEV</td>
<td>$q_{cl} \rightarrow q$</td>
<td>Evaporation of liquid cloud drops settling out of cloud</td>
<td>(1)</td>
</tr>
<tr>
<td>RACW</td>
<td>$q_{cl} \rightarrow q_{R}$</td>
<td>Collection of liquid cloud by rain</td>
<td></td>
</tr>
<tr>
<td>RAUT</td>
<td>$q_{cl} \rightarrow q_{R}$</td>
<td>Autoconversion of liquid cloud to rain</td>
<td></td>
</tr>
<tr>
<td>REVP</td>
<td>$q_{R} \rightarrow q$</td>
<td>Evaporation of rain</td>
<td></td>
</tr>
<tr>
<td>IACW</td>
<td>$q_{cl} \rightarrow q_{ic}$</td>
<td>Collection of liquid cloud by ice crystals</td>
<td></td>
</tr>
<tr>
<td>IDEP1</td>
<td>$q_{cl} \rightarrow q_{ic} \rightarrow q_{ic}$</td>
<td>Deposition of vapour from liquid cloud onto ice crystals</td>
<td>(3)</td>
</tr>
<tr>
<td>IDEP2</td>
<td>$q \rightarrow q_{ic}$</td>
<td>Deposition of vapour onto ice crystals</td>
<td></td>
</tr>
<tr>
<td>IPRM1</td>
<td>$q_{cl} \rightarrow q_{ic}$</td>
<td>Heterogeneous nucleation of ice from liquid cloud</td>
<td>(4)</td>
</tr>
<tr>
<td>IPRM2</td>
<td>$q \rightarrow q_{ic}$</td>
<td>Heterogeneous nucleation of ice from vapour</td>
<td>(4)</td>
</tr>
<tr>
<td>IFRW</td>
<td>$q_{cl} \rightarrow q_{ic}$</td>
<td>Homogeneous freezing of liquid cloud</td>
<td></td>
</tr>
<tr>
<td>IMLT</td>
<td>$q_{ic} \rightarrow q_{R}$</td>
<td>Melting of ice crystals</td>
<td></td>
</tr>
<tr>
<td>ISUB</td>
<td>$q_{ic} \rightarrow q$</td>
<td>Sublimation of ice crystals</td>
<td>(5)</td>
</tr>
<tr>
<td>IMLTEV</td>
<td>$q_{ic} \rightarrow q$</td>
<td>Evaporation of melting ice crystals</td>
<td>(5)</td>
</tr>
<tr>
<td>IACR</td>
<td>$q_{R} \rightarrow q_{ic}$</td>
<td>Collection of rain by ice crystals</td>
<td></td>
</tr>
<tr>
<td>SACW</td>
<td>$q_{cl} \rightarrow q_{icfa}$</td>
<td>Collection of liquid cloud by ice aggregates</td>
<td></td>
</tr>
<tr>
<td>SDEP1</td>
<td>$q_{cl} \rightarrow q_{icfa}$</td>
<td>Deposition of vapour from liquid cloud onto ice aggregates</td>
<td>(3)</td>
</tr>
<tr>
<td>SDEP2</td>
<td>$q \rightarrow q_{icfa}$</td>
<td>Deposition of vapour onto ice aggregates</td>
<td></td>
</tr>
<tr>
<td>SMLT</td>
<td>$q_{icfa} \rightarrow q_{R}$</td>
<td>Melting of ice aggregates</td>
<td></td>
</tr>
<tr>
<td>SSUB</td>
<td>$q_{icfa} \rightarrow q$</td>
<td>Sublimation of ice aggregates</td>
<td>(5)</td>
</tr>
<tr>
<td>SMLTEV</td>
<td>$q_{icfa} \rightarrow q$</td>
<td>Evaporation of melting ice aggregates</td>
<td>(5)</td>
</tr>
<tr>
<td>SACR</td>
<td>$q_{R} \rightarrow q_{icfa}$</td>
<td>Collection of ice crystals by aggregates</td>
<td></td>
</tr>
<tr>
<td>SAUT</td>
<td>$q_{ic} \rightarrow q_{icfa}$</td>
<td>Autoconversion of ice crystals to aggregates</td>
<td></td>
</tr>
<tr>
<td>SACI</td>
<td>$q_{ic} \rightarrow q_{icfa}$</td>
<td>Collection of ice crystals by aggregates</td>
<td></td>
</tr>
<tr>
<td>GAUT</td>
<td>$q_{icfa} \rightarrow q_{graup}$</td>
<td>Autoconversion of ice aggregates to graupel</td>
<td></td>
</tr>
<tr>
<td>GACW</td>
<td>$q_{icfa} \rightarrow q_{graup}$</td>
<td>Collection of liquid cloud by graupel</td>
<td></td>
</tr>
<tr>
<td>GACS</td>
<td>$q_{icfa} \rightarrow q_{graup}$</td>
<td>Collection of ice aggregates by graupel</td>
<td></td>
</tr>
<tr>
<td>GMLT</td>
<td>$q_{graup} \rightarrow q_{R}$</td>
<td>Melting of graupel</td>
<td></td>
</tr>
</tbody>
</table>

(1) LSETEV: Liquid cloud droplets falling into the clear-sky portion of a gridbox are evaporated across the whole size spectrum (however the falling droplets will be predominantly the larger ones with faster terminal velocities in the first place). Any contained chemical or aerosol species are released.

(2) REVP: Raindrops shrink by evaporation, causing chemical species to be released until effective Henry’s law equilibrium is restored. Aerosol is only released from those raindrops which evaporate completely, each of which produces one aerosol particle.

(3) IDEP1/SDEP1: Deposition occurs preferentially from the liquid phase, to represent the Bergeron-Findeisen process. However this involves evaporation followed immediately by deposition, thus any contained chemical or aerosol species are left behind in the liquid phase and released according to effective Henry’s law (for chemical species) or for completely-evaporated droplets (for aerosol).

(4) IPRM1/IPRM2: Heterogeneous nucleation involves the take-up of ice nuclei; however this is handled separately at the start of each timestep according to the scavenging coefficients.

(5) ISUB/IMLTEV/SSUB/SMLTEV: Ice particles shrink by sublimation or melting followed by evaporation. Aerosol is only released from those particles which sublimate (or melt and evaporate) completely, each of which produces one aerosol particle. Chemical species should re-equilibrate according to effective Henry’s law.
Any chemical or aerosol which is thus taken up into droplets or ice particles is excluded from the “dry” processes, in particular dry deposition and sedimentation (as they will instead fall along with the water). Aerosol produced by wet oxidation is assumed to be taken up into the cloud liquid water.

We have not discussed the take-up of chemical species by ice particles here – although such take-up has been observed, and a number of mechanisms proposed, there is no established theoretical framework to describe them (Pruppacher and Klett, 1996, §17.5.4); such processes are thus neglected for now.

When aerosol is returned to the prognostic tracers, either due to evaporation/sublimation or at the end of the timestep, it assumes a size distribution derived from the (assumed) size distribution of the droplets or ice particles in which it was contained. For evaporation/sublimation, we assume that each hydrometeor leaves behind one aerosol particle; however there are two complications which must be considered:

- If evaporation/sublimation is not complete, only the smallest hydrometeors will do so fully (thus releasing aerosol particles) while the larger particles will shrink (Pruppacher and Klett, 1996, §13.2.3). A theoretical calculation of the size distribution of hydrometeors which are fully evaporated/sublimated is not straightforward (see e.g. Tzivion et al., 1989); it thus seems prudent to carry out a sensitivity study with the limiting cases before doing so. Such cases could be (a) no hydrometeors shrink, and a uniform fraction across the size spectrum are removed; (b) no hydrometeors shrink, and only the smallest are removed; and (c) all hydrometeors shrink, and none are removed until no liquid or ice remains.

- The concentration of aerosol in hydrometeors (mass of aerosol per unit mass of water/ice) may not be uniform, but rather vary across the size spectrum. This is particularly so for small droplets, where growth is dominated by condensation and size well correlated with that of the initial condensation nucleus (Pruppacher and Klett, 1996, §17.3); for larger droplets growth is dominated by coalescence which leads to a more uniform concentration by the time raindrops are formed. In addition, the production of secondary aerosol material within the droplet by aqueous chemistry may not be proportional to the droplet mass, e.g. due to different dissolved concentrations of gaseous precursors; detailed modelling by Ovchinnikov and Easter (2010) suggests this can have a significant impact on the relationship between droplet size and dissolved aerosol concentration. A further sensitivity study may be needed to assess the effect of different assumptions for this relationship.

The scheme is summarised in Table 3.3.

3.3 Mathematical formulation

The microphysical transfer terms in Table 3.2 can be grouped into mass transfers between the different phases (these are all in mass of water per unit mass of air per unit time, i.e.
Table 3.3: Overview of a consistent in-cloud wet deposition scheme for UKCA.

<table>
<thead>
<tr>
<th>Chemistry</th>
<th>Aerosols</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 Calculate mass ratio of chemical species in cloud liquid water (and prognostic rain if enabled) using effective Henry’s law, based on actual liquid water content from cloud scheme.</td>
<td>Calculate number and mass ratio of aerosol species in cloud liquid water and ice (and prognostic rain/snow/graupel if enabled) using separate number/mass scavenging coefficients for each mode and phase (either fixed, or diagnosed from the activation scheme).</td>
</tr>
<tr>
<td>Chemical species diagnosed to be in an aqueous phase should be subject to aqueous-phase chemistry only, and not to gas-phase chemistry.</td>
<td>Aerosol diagnosed to be in an aqueous phase should not be subject to any of the dry aerosol processes.</td>
</tr>
<tr>
<td>Chemical species produced by aqueous-phase reactions should go directly into the diagnosed aqueous mass, rather than the gas-phase mass.</td>
<td>Aerosol produced by wet oxidation should go directly into the diagnosed aqueous mass, rather than the dry aerosol mass.</td>
</tr>
<tr>
<td>2 Iterating down the wet levels in a column, transfer mass between the different phases, and pass to the level below, in proportion to the water transferred, as given by the microphysics and fall-rate diagnostics from the precipitation scheme.</td>
<td>For water which returns to the vapour phase, add it to the dry aerosol number and mass, with a size distribution given by the assumed size distribution of the source phase.</td>
</tr>
<tr>
<td>Re-equilibrate falling drops to their new environment according to effective Henry’s law.</td>
<td>Update the number and mass tracers, giving aerosol in aqueous phases a size distribution according to that of the water phase in which it is dissolved.</td>
</tr>
</tbody>
</table>
| 3 Feed the net change in mass from the transfers into the solver. | }
3.3. MATHEMATICAL FORMULATION

\[ kg_{\text{water}} \text{ kg}^{-1} \text{ s}^{-1} \]:

\[
T_{q_{cl} \rightarrow q} = P_{\text{SETEV}} + P_{\text{DEP1}} \tag{3.1}
\]

\[
T_{q_{cl} \rightarrow q_{R}} = P_{\text{RAUT}} + P_{\text{RAUT}} \tag{3.2}
\]

\[
T_{q_{R} \rightarrow q} = P_{\text{REVP}} \tag{3.3}
\]

\[
T_{q_{cl} \rightarrow q_{cfa}} = P_{\text{IACW}} + P_{\text{PRM1}} + P_{\text{PRM2}} \tag{3.4}
\]

\[
T_{q \rightarrow q_{cfa}} = P_{\text{DEP1}} + P_{\text{DEP2}} + P_{\text{PRM2}} \tag{3.5}
\]

\[
T_{q_{cfa} \rightarrow q} = P_{\text{IMLT}} \tag{3.6}
\]

\[
T_{q_{cfa} \rightarrow q_{R}} = P_{\text{SUB}} + P_{\text{MLT}} \tag{3.7}
\]

\[
T_{q_{R} \rightarrow q_{cfa}} = P_{\text{ACR}} \tag{3.8}
\]

\[
T_{q_{cfa} \rightarrow q_{R}} = P_{\text{ACR}} \tag{3.9}
\]

\[
T_{q_{cfa} \rightarrow q_{R}} = P_{\text{ACR}} \tag{3.10}
\]

\[
T_{q_{fa} \rightarrow q_{R}} = P_{\text{SMLT}} \tag{3.11}
\]

\[
T_{q_{fa} \rightarrow q} = P_{\text{SUB}} + P_{\text{MLT}} \tag{3.12}
\]

\[
T_{R_{R} \rightarrow q_{fa}} = P_{\text{SACR}} \tag{3.13}
\]

\[
T_{q_{cfa} \rightarrow q_{fa}} = P_{\text{SACR}} \tag{3.14}
\]

\[
T_{q_{fa} \rightarrow q_{R}} = P_{\text{GAUT}} + P_{\text{GACS}} \tag{3.15}
\]

\[
T_{q_{R} \rightarrow q_{fa}} = P_{\text{GACW}} \tag{3.16}
\]

\[
T_{q_{fa} \rightarrow q_{R}} = P_{\text{GMLT}} \tag{3.17}
\]

The total mass mixing ratio \( m_{T}^{(i)} \) of a chemical species \( i \) is split into a gaseous portion \( m^{(i)} \) and portions taken up by each of the water phases \( (m_{cl}^{(i)}, m_{cfc}^{(i)}, m_{cfa}^{(i)}, m_{R}^{(i)}, m_{\text{graup}}^{(i)} ) \) so that

\[
m_{T}^{(i)} = m^{(i)} + m^{(i)} + m^{(i)} + m^{(i)} + m^{(i)} \tag{3.18}
\]

For aerosol species, the “free” or interstitial aerosol \( m^{(i)} \) is divided into modes \( m_{M}^{(i)} \), as in the existing scheme, while the portion taken up by the water phases follows the size distribution assumed for droplets or ice particles. Thus the same equation applies, with

\[
m^{(i)} = \sum_{M \in M} m_{M}^{(i)}, \tag{3.19}
\]

where \( M \) is the set of aerosol modes, i.e.

\[
m_{T}^{(i)} = m_{cl}^{(i)} + m_{cfc}^{(i)} + m_{cfa}^{(i)} + m_{R}^{(i)} + m_{\text{graup}}^{(i)} + \sum_{M \in M} m_{M}^{(i)} \tag{3.20}
\]

The evolution of water in each phase in the large-scale precipitation scheme is given by

\[
\frac{\partial q_{x}}{\partial t} = \sum_{y \in P} (T_{q_{y} \rightarrow q_{x}} - T_{q_{x} \rightarrow q_{y}}) + \frac{1}{\rho} \frac{\partial F_{x}}{\partial z} \tag{3.21}
\]

for each \( x \in P \), where \( P \) is the set of water phases and \( F_{x} \) the downward precipitation flux of water in that phase (an area flux, i.e. mass of water per unit horizontal area per unit time or kg m\(^{-2}\) s\(^{-1}\)).

The corresponding evolution of chemical or aerosol species in that water is given by

\[
\frac{\partial m_{x}^{(i)}}{\partial t} = S_{q_{x}} - R_{q_{x}}^{(i)} + \sum_{y \in P} \left( T_{q_{y} \rightarrow q_{x}} m_{y}^{(i)} - T_{q_{x} \rightarrow q_{y}} m_{x}^{(i)} \right) \frac{1}{\rho} \frac{\partial F_{x}}{\partial z} \left( q_{x} \right) m_{x}^{(i)} \tag{3.22}
\]
for each \( x \in \mathcal{P}' \), where \( \mathcal{P}' \) is the set of water phases excluding vapour, \( S_{q_x}^{(i)} \) is take-up (scavenging) from the gaseous/interstitial mode(s), and \( R_{q_x}^{(i)} \) is release to those modes.

Note that the \( T_{q_x} m_x^{(i)} / q_x \) terms are ill-defined where \( q_x \) approaches zero. However, where \( q_x \) is small, it is clear that the transfer of chemical/aerosol out of this phase must also be small. Thus to avoid division by zero, the terms are dropped altogether when \( q_x \) is below a threshold value.

For the evolution of chemical species in the gaseous state, or total interstitial aerosol across all modes,

\[
\frac{\partial m^{(i)}}{\partial t} = \sum_{x \in \mathcal{P}'} \left( R_{q_x}^{(i)} - S_{q_x}^{(i)} \right),
\]

(3.23)

For aerosol species, we must consider the size spectrum – \( S_{n_x}^{(i)}(r) \) or \( R_{n_x}^{(i)}(r) \) – of the aerosol taken up or released:

\[
\frac{\partial n^{(i)}(r)}{\partial t} = \sum_{x \in \mathcal{P}'} \left( R_{n_x}^{(i)}(r) - S_{n_x}^{(i)}(r) \right),
\]

(3.24)

with the total number and mass rates given by:

\[
S_{N_x}^{(i)} = \int_{0}^{\infty} S_{n_x}^{(i)}(r) \, dr \quad \quad S_{q_x}^{(i)} = \frac{4}{3} \pi \rho^{(i)} \int_{0}^{\infty} r^3 S_{n_x}^{(i)}(r) \, dr
\]

(3.25)

\[
R_{N_x}^{(i)} = \int_{0}^{\infty} R_{n_x}^{(i)}(r) \, dr \quad \quad R_{q_x}^{(i)} = \frac{4}{3} \pi \rho^{(i)} \int_{0}^{\infty} r^3 R_{n_x}^{(i)}(r) \, dr,
\]

(3.26)

assuming spherical aerosol particles. Given that at present we have only a one-moment bulk scheme for cloud and precipitation, this requires assumed size distributions for cloud water droplets, raindrops and ice particles. These are considered for each process in turn below, based on those used by the precipitation scheme itself (Wilkinson et al., 2009). In the future, it is likely that the Unified Model will gain a multi-moment prognostic cloud and precipitation scheme; when this is used, the predicted hydrometeor size distribution should be used instead.

### 3.3.1 Processes releasing aerosol from water phases to free/interstitial modes

**LSETEV: evaporation of settling droplets**

The implementation of droplet settling in the large-scale precipitation scheme assumes a Khrgian-Mazin gamma distribution \( \Gamma(3, \theta_{cl}) \) of droplet radii,

\[
n_{cl}(r) = \frac{N_{cl}}{2\theta_{cl}^3} r^2 e^{-\frac{r}{\theta_{cl}}},
\]

(3.27)

where \( N_{cl} = \int_{0}^{\infty} n_{cl}(r) \, dr \) is the total droplet number density (currently fixed values over land and sea, although it could be diagnosed via aerosol activation), and the parameter \( \theta_{cl} \) is given by

\[
\theta_{cl} = \sqrt[3]{\frac{\rho q_{cl}}{80 \pi \rho_w N_{cl}}}
\]

(3.28)

so that \( \rho q_{cl} = \frac{4}{3} \pi \rho_w \int_{0}^{\infty} r^3 n_{cl}(r) \, dr \) (Wilkinson et al., 2009, §6.2.1), where spherical droplets are assumed. The droplets are assumed to fall at a terminal speed of:

\[
V_{cl}(r) = K_1 r^2,
\]

(3.29)

where \( K_1 \) is a function of temperature only (Wilkinson et al., 2009, §6.2.1).
3.3. MATHEMATICAL FORMULATION

The downward number flux of droplets with radius \( r \) is thus

\[
F_{n_{\text{cl}}} (r) = n_{\text{cl}}(r) V_{\text{cl}}(r) = \frac{K_1 N_{\text{cl}}}{2 \theta_{\text{cl}}^3} r^4 e^{-\frac{r}{\theta_{\text{cl}}}},
\]

(3.30)

giving a total downward mass flux

\[
F_{q_{\text{cl}}} \downarrow = \frac{4}{3} \pi \rho_w \int_0^{\infty} r^3 F_{n_{\text{cl}}} (r) \, dr
\]

(3.31)

\[
= \frac{4}{3} \pi \rho_w \int_0^{\infty} K_1 N_{\text{cl}} r^7 e^{-\frac{r}{\theta_{\text{cl}}}} \, dr
\]

(3.32)

\[
= 42 K_1 \theta_{\text{cl}}^2 \rho_q_{\text{cl}}.
\]

(3.33)

Assuming that the distribution of droplets falling into cloudy and clear air are identical, the number rate of evaporation for droplets of radius \( r \) is

\[
T_{n_{\text{cl}} \rightarrow q}\big|_{\text{LSETEV}} = \left. \frac{T_{q_{\text{cl}} \rightarrow q}\big|_{\text{LSETEV}}}{F_{n_{\text{cl}}} (r)} \right|_{r_{\text{cl}}},
\]

(3.35)

\[
= \left. \frac{T_{q_{\text{cl}} \rightarrow q}\big|_{\text{LSETEV}}}{F_{n_{\text{cl}}} (r)} \right|_{\left. \frac{K_1 N_{\text{cl}}}{2 \theta_{\text{cl}}^3} r^4 e^{-\frac{r}{\theta_{\text{cl}}}} \right|_{r_{\text{cl}}}},
\]

(3.36)

\[
= \frac{N_{\text{cl}} T_{q_{\text{cl}} \rightarrow q}\big|_{\text{LSETEV}}}{84 \theta_{\text{cl}}^3 \rho_q_{\text{cl}}} r_{\text{cl}}^4 e^{-\frac{r_{\text{cl}}}{\theta_{\text{cl}}}},
\]

(3.37)

i.e. a \( \Gamma(5, \theta_{\text{cl}}) \) distribution with a total number rate

\[
T_{N_{\text{cl}} \rightarrow q}\big|_{\text{LSETEV}} = \frac{2 N_{\text{cl}} T_{q_{\text{cl}} \rightarrow q}\big|_{\text{LSETEV}}}{7 \rho_q_{\text{cl}}},
\]

(3.38)

Assuming a uniform concentration of the aerosol species \( i \) in the cloud water (which, as noted earlier, may not be realistic for small droplets), a spherical droplet of radius \( r_{\text{cl}} \) leaves behind a spherical aerosol particle of dry radius

\[
r^{(i)} = \phi_{\text{cl}}^{(i)} r_{\text{cl}},
\]

(3.39)

where

\[
\phi_{\text{cl}}^{(i)} = \sqrt{\frac{\rho_w m_{\text{cl}}^{(i)}}{\rho^{(i)} q_{\text{cl}}}},
\]

(3.40)

so that the dry size distribution of released aerosol particles is given by

\[
R_{n_{\text{cl}}}^{(i)}\big|_{\text{LSETEV}} (r^{(i)}) \, dr^{(i)} = T_{n_{\text{cl}} \rightarrow q}\big|_{\text{LSETEV}} (r_{\text{cl}}) \, dr_{\text{cl}},
\]

(3.41)

which leads to a \( \Gamma(5, \theta_{\text{cl}}^{(i)}) \) distribution:

\[
P_{n_{\text{cl}}}^{(i)}\big|_{\text{LSETEV}} (r^{(i)}) = \left. \frac{T_{q_{\text{cl}} \rightarrow q}\big|_{\text{LSETEV}}}{F_{n_{\text{cl}}} (r)} \right|_{\left. \frac{K_1 N_{\text{cl}} \theta_{\text{cl}}^2}{2 \left( \theta_{\text{cl}}^{(i)} \right)^5} r^{(i)} e^{-\frac{r^{(i)}}{\theta_{\text{cl}}^{(i)}}} \right|_{r_{\text{cl}}}},
\]

(3.42)

\[
= \frac{N_{\text{cl}} T_{q_{\text{cl}} \rightarrow q}\big|_{\text{LSETEV}}}{84 \rho_q_{\text{cl}} \left( \theta_{\text{cl}}^{(i)} \right)^5} \left( r^{(i)} \right)^4 e^{-\frac{r^{(i)}}{\theta_{\text{cl}}^{(i)}}},
\]

(3.43)
where
\[ \theta_{cl}^{(i)} = \theta_{cl}^{(i)} \]
\[ = \sqrt[3]{\frac{\rho m^{(i)}_{cl}}{80\pi N_{cl}^{(i)}}} \] \hspace{1cm} (3.44)
\[ = 3\sqrt{\frac{\rho m^{(i)}_{cl}}{80\pi N_{cl}^{(i)}}} \]
\[ \phi^{(i)}_{cl} \]
\[ = 3\sqrt{\frac{\rho m^{(i)}_{cl}}{80\pi N_{cl}^{(i)}}} \]
\[ \times \frac{80\pi N_{cl}^{(i)}}{\rho^{(i)}_{cl}} \times \frac{1}{\rho^{(i)}_{cl}} \] \hspace{1cm} (3.45)

This gives a total number rate
\[ R_{N_{cl}}^{(i)} \] \hspace{1cm} (3.46)
\[ = 2N_{cl} \frac{T_{q_{cl} \rightarrow q_{cl}}} { \rho_{q_{cl}}^{(i)}} \]
\[ \frac{\rho_{q_{cl}}^{(i)}}{q_{cl}} \] \hspace{1cm} (3.47)

and mass rate
\[ R_{q_{cl}}^{(i)} \] \hspace{1cm} (3.48)
\[ = \frac{m^{(i)}_{cl}} {q_{cl}} \frac{T_{q_{cl} \rightarrow q_{cl}}} {\rho_{q_{cl}}^{(i)}} \]

**REVP: evaporation of rain**

The large-scale precipitation scheme assumes a Marshall-Palmer negative-exponential distribution \( \text{Exp}(\lambda_R) \equiv \Gamma(1, \frac{1}{\lambda_R}) \) for raindrop diameters\(^1\).

\[ n_{R}(D) = n_{a,R} \lambda_R^{n_{b,R}} e^{-\lambda_R D} \] \hspace{1cm} (3.49)

where \( n_{a,R} \) and \( n_{b,R} \) are fixed parameters (note that \( n_{b,R} \) is a non-integer exponent), and \( \lambda_R \) will be determined from the rainfall rate (Wilkinson et al., 2009, §4.1).

The raindrops are assumed to fall at a terminal speed of:
\[ V_{R}(D) = c_{R} D^{d_{R}} \left( \frac{\rho_0}{\rho} \right) \frac{\varphi_{R}}{} \] \hspace{1cm} (3.50)

where \( c_{R}, d_{R}, \varphi_{R} \) and \( \rho_0 \) are fixed parameters (Wilkinson et al., 2009, §4.4); note that the exponents are non-integer. In fact, break-up of large drops is thought to lead to a significant number of smaller drops travelling at super-terminal speed due to their finite relaxation time (Montero-Martínez et al., 2009); however we follow assumption of the precipitation scheme here.

The downward number flux of raindrops with diameter \( D \) is thus
\[ F_{n_{R} \downarrow}(D) = n_{R}(D) V_{R}(D) \] \hspace{1cm} (3.51)
\[ = n_{a,R} c_{R} \left( \frac{\rho_0}{\rho} \right) \frac{\varphi_{R}}{} \lambda_R^{n_{b,R}} D^{d_{R}} e^{-\lambda_R D} , \]

where the parameter \( \lambda_R \) is given by
\[ \lambda_R = \left( \frac{\pi c_{R} \left( \frac{\rho_0}{\rho} \right) \varphi_{R}}{1 \rho_{w} n_{a,R}} \right)^{\frac{1}{\frac{4\pi d_{R}}{n_{b,R}}}} \] \hspace{1cm} (3.52)

so that \( F_{q_{R} \downarrow} = \frac{\pi}{6} \rho_{w} \int D^{3} n_{R}(D) V_{R}(D) \) dD (spherical raindrops are assumed).
3.3. MATHEMATICAL FORMULATION

Under the first of our proposed assumptions for the size-dependent evaporation of raindrops (no droplets shrink, and a uniform proportion across the size spectrum are removed), the complete-evaporation number rate for raindrops of diameter \( D \) is given by

\[
T_{n_R \rightarrow q}(D)|_{\text{REVP}} = \frac{T_{q_R \rightarrow q}|_{\text{REVP}} F_{n_R}(D)}{F_{q_R}(D)}
\]

and thus follows a \( \Gamma(d_R + 1, \lambda_R) \) distribution with a total number rate

\[
T_{n_R \rightarrow q}|_{\text{REVP}} = \frac{\Gamma(d_R + 1) T_{q_R \rightarrow q}|_{\text{REVP}} n_{a,R} c_R \left( \frac{\rho_0}{\rho} \right) \frac{\varphi_R}{\lambda_R} \lambda_{n_R,R}^{d_R \rho_R - \lambda_R D} \cdot (3.55)
\]

Assuming a uniform concentration of the aerosol species \( i \) in the rain water (not necessarily realistic, as noted earlier, but probably more so than for cloud droplets since growth to raindrop size is dominated by coalescence), a spherical droplet of diameter \( D_R \) leaves behind a spherical aerosol particle of dry radius

\[
r^{(i)} = \frac{1}{2} \varphi_R^{(i)} D_R,
\]

so that the dry size distribution of released aerosol particles is given by

\[
R^{(i)}_{n_R}|_{\text{REVP}} (r^{(i)}) \, \text{d}r^{(i)} = T_{n_R \rightarrow q}|_{\text{REVP}} (D_R) \, \text{d}D_R,
\]

which leads to a \( \Gamma(d_R + 1, \lambda_R) \) distribution:

\[
R^{(i)}_{n_R}|_{\text{REVP}} (r^{(i)}) = \frac{T_{q_R \rightarrow q}|_{\text{REVP}}}{F_{q_R}(D_R)} \left( \frac{\rho_0}{\rho} \right) \frac{\varphi_R}{\lambda_R} \lambda_{n_R,R}^{d_R \rho_R - \lambda_R D} \cdot (3.60)
\]

where

\[
\lambda_R^{(i)} = \lambda_R \frac{2}{\varphi_R}
\]

This gives a total number rate

\[
R^{(i)}_{n_R}|_{\text{REVP}} = T_{n_R \rightarrow q}|_{\text{REVP}} \cdot (3.63)
\]

and mass rate

\[
R^{(i)}_{q_R}|_{\text{REVP}} = \frac{m^{(i)}_{q_R}}{q_R} T_{q_R \rightarrow q}|_{\text{REVP}} \cdot (3.65)
\]
I Dep1/S Dep1: Bergeron-Findeisen process

When liquid water is converted to ice via the Bergeron-Findeisen process, it is first evaporated before being deposited onto existing ice particles. As for droplet settling, we assume a Khrgjan-Mazin gamma distribution $\Gamma(3, \theta_{cl})$,

$$n_{cl}(r) = \frac{N_{cl}}{2\theta_{cl}^3} r^2 e^{-\frac{r}{\theta_{cl}}}.$$  \hfill (3.66)

If we again assume uniform total evaporation across the size spectrum, the number rate of evaporation for droplets of diameter $D$ is

$$T_{n_{cl} \rightarrow q}(r)|_{I Dep1+S Dep1} = \frac{n_{cl}(r)}{q_{cl}} T_{q_{cl} \rightarrow q}|_{I Dep1+S Dep1}$$  \hfill (3.67)

$$= \frac{N_{cl} T_{q_{cl} \rightarrow q}|_{I Dep1+S Dep1}}{2\theta_{cl}^3 q_{cl}} r^2 e^{-\frac{r}{\theta_{cl}}}.$$  \hfill (3.68)

with a total number rate

$$T_{N_{cl} \rightarrow q}|_{I Dep1+S Dep1} = \frac{N_{cl} T_{q_{cl} \rightarrow q}|_{I Dep1+S Dep1}}{q_{cl}},$$  \hfill (3.69)

so that the dry size distribution of released aerosol particles is given by

$$R((i))_{n_{cl} | I Dep1+S Dep1} = N_{cl} T_{q_{cl} \rightarrow q}|_{I Dep1+S Dep1} (r_{cl}) \frac{2}{2q_{cl} (\theta_{cl}(i))^3} e^{-\frac{r_{cl}(i)}{\theta_{cl}(i)}},$$  \hfill (3.71)

with a total number rate

$$R((i))_{N_{cl} | I Dep1+S Dep1} = T_{N_{cl} \rightarrow q}|_{I Dep1+S Dep1}$$  \hfill (3.72)

$$= \frac{N_{cl} T_{q_{cl} \rightarrow q}|_{I Dep1+S Dep1}}{q_{cl}},$$  \hfill (3.73)

and mass rate

$$R((i))_{q_{cl} | I Dep1+S Dep1} = \frac{m((i))_{cl}}{q_{cl}} T_{q_{cl} \rightarrow q}|_{I Dep1+S Dep1}.$$  \hfill (3.74)

ISub/IM LTEV/SSub/SMLTEV: sublimation or evaporation of melting ice

The treatment of ice particles is made more complex by the fact that they are not in general spherical. The mass-diameter relationship is assumed to take the form of a power law, with the mass of a particle in phase $x$ given by

$$M_x(D) = a_x D^{b_x},$$  \hfill (3.75)

where $a_x$ and $b_x$ are fixed parameters\(^2\) (Wilkinson et al., 2009, §4.5). Particle diameters are assumed to follow a $\Gamma(\alpha_x + 1, \frac{1}{\lambda_x})$ distribution, with the number of particles of diameter $D$ given by

$$n_x(D) = n_{a,x} \lambda_x^{n_x} D^{\alpha_x} e^{-\lambda_x D},$$  \hfill (3.76)

\(^2\)This is a generalisation of the relationship for spherical liquid water drops (which are assumed for liquid cloud and rain), where $a_{cl} = a_{l1} = \frac{4}{3} \times 10^3 \text{kg} \text{m}^{-3}$ and $b_{l1} = b_{cl} = 3$.}
where \( n_{a,x}, n_{b,x} \) and \( \alpha_x \) are fixed parameters (Wilkinson et al., 2009, §4.1), and \( \lambda_x \) is given by

\[
\lambda_x = \left( \frac{\Gamma (\alpha_x + b_x + 1) n_{a,x} a_x}{q_x} \right)^{\frac{1}{\alpha_x + b_x + n_{b,x}}} \tag{3.77}
\]

so that \( q_x = \int_0^\infty a_x D^{b_x} n_x(D) \, dD \). As for rain, the fall velocities are given by

\[
V_x(D) = c_x D^{d_x} \left( \frac{\rho_0}{\rho} \right) ^{\frac{q_x}{G_x}}, \tag{3.78}
\]

where \( c_x, d_x, G_x \) and \( \rho_0 \) are fixed parameters.

The downward number flux of ice particles with diameter \( D \) is thus

\[
F_{n_x,1}(D) = n_x(D) V_x(D) \tag{3.79}
\]

\[
= n_{a,x} c_x \left( \frac{\rho_0}{\rho} \right) ^{\frac{q_x}{G_x}} \lambda_x^{n_{b,x}} D^{\alpha_x + d_x} e^{-\lambda_x D}. \tag{3.80}
\]

Instead of determining \( \lambda_x \) from the ice water content as above, we could also determine it from the snowfall rate as

\[
\lambda_x = \left( \frac{\Gamma (\alpha_x + b_x + 1) n_{a,x} a_x c_x}{F_{q_x,1}} \right)^{\frac{1}{1+\alpha_x + b_x + n_{b,x}}} \tag{3.81}
\]

so that \( F_{q_x,1} = \int_0^\infty a_x D^{b_x} n_x(D) V_x(D) \, dD \).

Under the assumption that a uniform proportion of ice particles across the size spectrum sublimes (or melts and evaporates) completely, the number rate for particles of diameter \( D \) is given by

\[
T_{n_x \to q}(D)|_P = \frac{T_{q_x \to q}|_P F_{n_x,1}(D)}{F_{q_x,1}} \tag{3.82}
\]

\[
= \frac{T_{q_x \to q}|_P}{F_{q_x,1}} n_{a,x} c_x \left( \frac{\rho_0}{\rho} \right) ^{\frac{q_x}{G_x}} \lambda_x^{n_{b,x}} D^{\alpha_x + d_x} e^{-\lambda_x D}, \tag{3.83}
\]

where \( P \in \{ \text{ISUB, SSUB, IMLTEV, SMLTEV} \} \), and thus follows \( \Gamma (\alpha_x + d_x + 1, \frac{1}{\lambda_x}) \) distribution with a total number rate

\[
T_{N_x \to q}|_P = \frac{\Gamma (\alpha_x + d_x + 1) T_{q_x \to q}|_P}{\Gamma (\alpha_x + b_x + d_x + 1)} \left( \frac{\Gamma (\alpha_x + b_x + d_x + 1) n_{a,x} c_x}{F_{q_x,1}} \left( \frac{\rho_0}{\rho} \right) ^{\frac{q_x}{G_x}} \right)^{\frac{b_x}{1+\alpha_x + b_x + n_{b,x}}}. \tag{3.84}
\]

Assuming a uniform concentration of the aerosol species \( i \) in the ice, an ice particle of diameter \( D_x \) leaves behind a spherical aerosol particle of dry radius

\[
r^{(i)} = \frac{1}{2} \frac{\phi^{(i)}_{x}}{D_R^{b_x}} \tag{3.85}
\]

where

\[
\phi^{(i)}_{x} = \sqrt{\frac{6 a_x m^{(i)}_{x}}{\pi^3 \rho^{(i)} q_x}}, \tag{3.86}
\]

so that the dry size distribution of released aerosol particles is given by

\[
R_{n_x \to q}|_P (r^{(i)}) \, dr^{(i)} = T_{n_x \to q}|_P (D_x) \, dD_x, \tag{3.87}
\]
which leads to a generalised gamma distribution:

$$R_n(x) = \frac{2}{\phi_x} \left( \frac{\rho_0}{\rho} \right) \frac{\rho_0}{\rho} \phi_x \frac{2}{\phi_x} (1+\alpha_x+d_x)^{-\alpha_x-d_x} n_{a,x} c_x \frac{\lambda^{(i)}_x}{\rho_0} \frac{3}{\phi_x} x (1+\alpha_x+d_x)^{-\alpha_x-d_x} n_{a,x} c_x \frac{\lambda^{(i)}_x}{\rho_0} \frac{3}{\phi_x} x,$$

with a total number rate

$$R_n = T_n \frac{n_i}{p},$$

and mass rate

$$R_q = m_i q x T_n \frac{n_i}{p},$$

where

$$\lambda^{(i)}_x = \frac{2}{\phi_x} \left( \frac{\rho_0}{\rho} \right)^{\frac{b_x}{3}} \left( \frac{\rho_0}{\rho} \right)^{\frac{b_x}{3}}.$$

### 3.3.2 Processes taking up free/interstitial aerosol to water phases

#### Activation and heterogeneous nucleation

Because there is no prognostic treatment of the aerosol taken up by cloud water and ice, the cumulative take-up of aerosol due to activation of CCN to form liquid cloud droplets and heterogeneous nucleation of ice must be diagnosed at the start of each time-step.

In the scheme currently used in UKCA-MODE, a fixed fraction $R_{scav,M}$ of the mass and number in each mode $M$ is assumed to be activated within the cloudy fraction of each grid box. This is not physically realistic however, as (for liquid cloud at least) Köhler theory predicts that particles will activate above a critical radius depending on their composition and the local supersaturation; thus large particles are preferentially activated and a higher proportion of mass than number will be activated.

The Abdul-Razzak and Ghan (2000) activation scheme, currently being introduced into the model by Rosalind West, diagnoses the critical radius $r_{crit,M}$ for each mode based on its composition and the estimated maximum local supersaturation.

In either case, the number of aerosol particles taken up into liquid cloud water is

$$n_n(r) = \begin{cases} C_x n_i(r) & r > r_{crit}^{(i)} \\ 0 & r \leq r_{crit}^{(i)} \end{cases}$$

where $n_i(r)$ is the prognostic aerosol number, and thus the total aerosol mass taken up is

$$m_n = \frac{4\pi \rho^{(i)}}{3} \int_{r_{crit}^{(i)}}^{\infty} r^3 n_n(r) dr.$$
scheme akin to that for activation would be desirable. Additional effects in mixed-phase clouds will need to be considered: it may be that, for example, soluble particles can be scavenged into ice in mixed-phase clouds but not in ice-only clouds – they may be activated in cloud droplets which then freeze, which cannot be “remembered” without prognostic in-cloud aerosol.

Once the scavenged aerosol has been calculated, the remaining (interstitial) aerosol number and mass are then

\[ n^{(i)}(r) = n^{(i)}_T(r) - \sum_{x \in P'} n^{(i)}_x(r) \] (3.95)

\[ m^{(i)}(r) = m^{(i)}_T(r) - \sum_{x \in P'} m^{(i)}_x(r). \] (3.96)

**Impaction scavenging**

The existing scheme for impaction scavenging by rain can be retained, but will be incorporated into the framework described here, calculating the \( S^{(i)}_{n_x}(r) \) term. In this way, aerosol taken up by impaction scavenging may be re-released at a lower level if the rain evaporates. Consideration should be given to extending this to cover impaction scavenging by liquid cloud droplets (which may lead to cloud processing of particles too small to activate even in non-precipitating clouds) and ice particles, giving the other \( S^{(i)}_{n_x}(r) \) terms.

### 3.3.3 Implementation in modal aerosol scheme

The aerosol size spectrum is represented in the model by a superposition of log-normal modes, with fixed (geometric) standard deviation and variable mean (within ranges separated by an order of magnitude).

If we assume that the size spectrum \( R^{(i)}_{n_x}(r)|_P \) of aerosol released by a given evaporation or sublimation process \( P \) is “narrow” compared to the order-of-magnitude separation between the log-normal modes, it is reasonable to simply transfer the total number \( R^{(i)}_{n_x}|_P \) and mass \( R^{(i)}_{q_x}|_P \) released into the mode \( M \) which contains the mean-volume radius

\[ \bar{r}_P = \left( \frac{3 R^{(i)}_{q_x}|_P}{4 \pi \rho^{(i)} R^{(i)}_{n_x}|_P} \right)^{\frac{1}{3}} \] (3.97)

of the released aerosol:

\[ \left. \frac{\partial N^{(i)}_M}{\partial t} \right|_P = R^{(i)}_{n_x}|_P \]

\[ \left. \frac{\partial m^{(i)}_M}{\partial t} \right|_P = R^{(i)}_{q_x}|_P. \] (3.98)

In this way, we avoid having to explicitly integrate over the whole size distribution.

For a gamma-distributed variable \( X \sim \Gamma(k, \theta) \), its standard deviation is

\[ \sqrt{\text{Var}(X)} = \frac{\mathbb{E}(X)}{\sqrt{k}}, \] (3.99)

and so, provided \( k \gg 1 \), this should be a reasonable assumption for modes separated by an order of magnitude.
CHAPTER 3. MICROPHYSICALLY-CONSISTENT SCAVENGING

Following this approach, the equations for modal aerosol become:

\[
\frac{\partial N^{(i)}_M}{\partial t} = - \sum_{x \in P} I^{(i)}_{n_x}(M) + \sum_{P \in E, r_P \in M} R^{(i)}_{N_x} \bigg|_P \quad (M \in M) \tag{3.100}
\]

\[
\frac{\partial m^{(i)}_M}{\partial t} = - \sum_{x \in P} I^{(i)}_{m_x}(M) + \sum_{P \in E} \bar{r}_P \in M R^{(i)}_{q_x} \bigg|_P \quad (M \in M) \tag{3.101}
\]

\[
\frac{\partial m^{(i)}_x}{\partial t} = \sum_{M \in M} I^{(i)}_{m_x}(M) - \sum_{P \in E} R^{(i)}_{q_x} \bigg|_P + \sum_{y \in P'} \left( T_{q_y \rightarrow q_x} m^{(i)}_y q_y - T_{q_x \rightarrow q_y} m^{(i)}_x q_x \right) + \frac{1}{\rho} \frac{\partial}{\partial z} \left( F_{q_x} m^{(i)}_x q_x \right), \tag{3.102}
\]

where \( E = \{ \text{LSETEV}, \text{REVP}, \text{IDEP1}, \text{SDEP1}, \text{ISUB}, \text{SSUB}, \text{IMLTEV}, \text{SMLTEV} \} \) is the set of evaporation/sublimation processes and \( I^{(i)}_{n_x}(M) \) and \( I^{(i)}_{m_x}(M) \) are the number and mass scavenged from mode \( M \) by impaction with water phase \( x \), as diagnosed by the impaction scavenging scheme.

A nucleation scavenging (activation) term does not appear in the time-evolution equations, as it is diagnosed as an “instantaneous” process at the start of the timestep (see Section 3.3.2). (In a refined scheme, we may wish to consider separately in these equations the activation which occurs during the timestep, as distinct from that assumed to have already occurred.)

For modal aerosol, the total activated number and mass in each mode can be calculated by integrating (3.93) and (3.94) over the lognormal distribution \( n^{(i)}_M(r) \):

\[
N^{(i)}_{M, cl} = \int_{r_{\text{crit}, M}}^{\infty} n^{(i)}_M(r) \, dr \tag{3.103}
\]

\[
N^{(i)}_{M, cl} = \frac{4\pi \rho^{(i)}_M}{3} \int_{r_{\text{crit}, M}}^{\infty} n^{(i)}_M(r) \, dr, \tag{3.104}
\]

calculating these as in the existing mode-merging algorithm.

### 3.4 Discretisation

The large-scale precipitation scheme solves the transfer processes sequentially; for consistency the chemical/aerosol transfers for each process \( P \) should be calculated in the same sequence, using the values of \( q_x \) on input to the corresponding process in the precipitation scheme. These mid-timestep values, denoted \( q_x|_P \), can be determined by re-applying the diagnosed transfer rates in the same sequence as the original calculations:

\[
q_x|_{P_{j+1}} = q_x|_{P_j} + \frac{\partial q_x}{\partial t} \bigg|_{P_j} \Delta t \quad (j \in \{0, 1, \ldots, J-1\}) \tag{3.105}
\]

(Note that this cannot be done correctly in an operator-split manner if iterated microphysics are in use, without additional diagnostics for the per-iteration transfer rates.)
The full discrete equation set for modal aerosol species is given below, based on the continuous versions derived in the preceding sections. In what follows, \( T_{X \rightarrow Y} \) and \( F_{X} \) are the sets of processes contributing to the transfer \( T_{X \rightarrow Y} \) or flux \( F_{X} \) respectively, \( F_{q_{z}} \) is the downward area flux of \( q_{z} \) across the top boundary of the grid box (equal to \( F_{q_{z}} \) in the box above), and \( m_{x}^{(i)} \) and \( q_{x} \) represent the values of \( m_{x}^{(i)} \) and \( q_{x} \) in the box above.

At the start of the timestep, in-cloud aerosol is split out diagnostically by the activation scheme (see Section 3.3.2):

\[
N_{M}^{(i)} = N_{M,T}^{(i)} - \sum_{x \in P'} N_{M,x}^{(i)}, \quad (3.106)
\]

\[
m_{M}^{(i)} = m_{M,T}^{(i)} - \sum_{x \in P'} m_{M,x}^{(i)}. \quad (3.107)
\]

The increments to mass and number in each mode from each microphysical process \( P \) in the large-scale precipitation scheme are

\[
\Delta N_{M}^{(i)} \big|_{P} = R_{N_{a}}^{(i)} \big|_{P} \Delta t \quad (M \in M, P \in \mathcal{E}, \bar{r}_{P} \in M), \quad (3.108)
\]

\[
\Delta m_{M}^{(i)} \big|_{P} = R_{q_{x}}^{(i)} \big|_{P} \Delta t \quad (M \in M, P \in \mathcal{E}, \bar{r}_{P} \in M), \quad (3.109)
\]

and the increments to in-cloud aerosol mass are given by

\[
\Delta m_{x}^{(i)} \big|_{P} = \begin{cases} 
\left( \frac{F_{q_{z}}^{(i)} m_{x}^{(i)} - F_{q_{z}}^{(i)} m_{x}^{(i)} |_{P}}{q_{z}^{(i)} |_{P}} \right) \Delta t / X & (P \in F_{q_{z}},) \\
\frac{P_{p} m_{a}^{(i)} |_{P} \Delta t}{q_{z}^{(i)} |_{P}} & (P \in T_{q_{z} \rightarrow q_{z}}, y \in P') \\
\frac{P_{p} m_{a}^{(i)} |_{P} \Delta t}{q_{z}^{(i)} |_{P}} & (P \in T_{q_{z} \rightarrow q_{z}}, y \in P') 
\end{cases} \quad (x \in P'), \quad (3.110)
\]

for falling/settling processes, evaporation/sublimation, and transfers into and out of the phase \( q_{z} \) respectively. (The expressions for \( R_{X} \) for the individual evaporation/sublimation processes are given at the end of this summary.)

The mid-timestep values of number and mass in each mode, and in-cloud mass, used for calculating these increments sequentially as per the precipitation scheme, are calculated as:

\[
N_{M}^{(i)} \big|_{P_{j} + 1} = \begin{cases} 
N_{M}^{(i)} \big|_{P_{j}} + \Delta N_{M}^{(i)} \big|_{P_{j}} & (P_{j} \in \mathcal{E}, \bar{r}_{P_{j}} \in M) \\
N_{M}^{(i)} \big|_{P_{j}} & \text{otherwise} 
\end{cases} \quad (M \in M, j \in \{0, 1, \ldots, J - 1\}), \quad (3.111)
\]

\[
m_{M}^{(i)} \big|_{P_{j} + 1} = \begin{cases} 
m_{M}^{(i)} \big|_{P_{j}} + \Delta m_{M}^{(i)} \big|_{P_{j}} & (P_{j} \in \mathcal{E}, \bar{r}_{P_{j}} \in M) \\
m_{M}^{(i)} \big|_{P_{j}} & \text{otherwise} 
\end{cases} \quad (M \in M, j \in \{0, 1, \ldots, J - 1\}), \quad (3.112)
\]

\[
m_{x}^{(i)} \big|_{P_{j} + 1} = \begin{cases} 
m_{x}^{(i)} \big|_{P_{j}} + \Delta m_{x}^{(i)} \big|_{P_{j}} & (P_{j} \in T_{q_{z}}) \\
m_{x}^{(i)} \big|_{P_{j}} & \text{otherwise} 
\end{cases} \quad (x \in P', j \in \{0, 1, \ldots, J - 1\}), \quad (3.113)
\]
where

\[
T_{q_x}^* = \left( \bigcup_{y \in P'} T_{q_y \rightarrow q_x} \right) \cup \left( \bigcup_{y \in P} T_{q_x \rightarrow q_y} \right) \tag{3.114}
\]

is the set of all transfer processes affecting the amount of aerosol in phase \(q_x\).

The total increments due to scavenging over the timestep are given by summing over both the contributions from precipitation microphysics and those from impaction scavenging (as noted before, activation is treated instantaneously at the start of the timestep, so does not appear here):

\[
\Delta N^{(i)}_M = \sum_{x \in P'} I^{(i)}_{n_x} (M) \Delta t + \sum_{P \in \mathcal{E}} \Delta N^{(i)}_M \bigg|_P \quad (M \in \mathcal{M}) \tag{3.115}
\]

\[
\Delta m^{(i)}_M = \sum_{x \in P'} I^{(i)}_{m_x} (M) \Delta t + \sum_{P \in \mathcal{T}^{q_x}} \Delta m^{(i)}_M \bigg|_P \quad (M \in \mathcal{M}) \tag{3.116}
\]

\[
\Delta m^{(i)}_x = \sum_{M \in \mathcal{M}} I^{(i)}_{m_x} (M) \Delta t + \sum_{P \in \mathcal{T}^{q_x}} \Delta m^{(i)}_M \bigg|_P \quad (x \in \mathcal{P}) \tag{3.117}
\]

Finally, the \(R_X\) terms for the number and mass of aerosol released by evaporation/sublimation processes are given below, as derived in Section 3.3.1. For evaporation of settling cloud droplets,

\[
R^{(i)}_{N_{cl}} \bigg|_{\text{LSETEV}} = \frac{2 N_{cl} \big|_{\text{LSETEV}} P_{\text{LSETEV}}}{\rho \ q_{cl} \big|_{\text{LSETEV}}} \tag{3.118}
\]

\[
R^{(i)}_{q_{cl}} \bigg|_{\text{LSETEV}} = \frac{P_{\text{LSETEV}} m^{(i)}_{cl} \big|_{\text{LSETEV}}}{q_{cl} \big|_{\text{LSETEV}}};
\]

and for evaporation of rain,

\[
R^{(i)}_{N_{R}} \bigg|_{\text{REVP}} = \frac{\Gamma (d_R + 1) P_{\text{REVP}}}{\pi \rho_w} \left( \frac{\pi c_R \left( \frac{\rho_0}{\rho} \right)}{F_{gr \downarrow}} \right)^{\frac{3}{1 + d_R - n_{b,R}}} \tag{3.119}
\]

\[
R^{(i)}_{q_{R}} \bigg|_{\text{REVP}} = \frac{m^{(i)}_{R} P_{\text{REVP}}}{q_{R}}.
\]

For the Bergeron-Findeisen process (deposition of evaporating droplets onto ice particles),

\[
R^{(i)}_{N_{cl}} \bigg|_{P} = \frac{N_{cl} P_{P}}{q_{cl}} \quad (P \in \{\text{IDEP1, SDEP1}\}); \tag{3.120}
\]

\[
R^{(i)}_{q_{cl}} \bigg|_{P} = \frac{m^{(i)}_{cl} P_{P}}{q_{cl}} \quad (P \in \{\text{IDEP1, SDEP1}\});
\]

for sublimation (or evaporation of melting) ice crystals,

\[
R^{(i)}_{N_{L_{cl}}} \bigg|_{P} = \frac{\Gamma (\alpha_{L_{cl}} + d_{L_{cl}} + 1) T_{L_{cl} \rightarrow q} \bigg|_{P}}{\Gamma (\alpha_{L_{cl}} + b_{L_{cl}} + d_{L_{cl}} + 1)} \times \left( \frac{\Gamma (\alpha_{L_{cl}} + b_{L_{cl}} + d_{L_{cl}} + 1) n_{a,L_{cl} \cdot L_{cl}}}{F_{L_{cl} \downarrow}} \right)^{\frac{b_{L_{cl}}}{1 + \alpha_{L_{cl}} + b_{L_{cl}} + d_{L_{cl}} - n_{b,L_{cl}}}} \tag{3.121}
\]

\[
R^{(i)}_{q_{L_{cl}}} \bigg|_{P} = \frac{m^{(i)}_{L_{cl}} T_{L_{cl} \rightarrow q} \bigg|_{P}}{q_{L_{cl}}}; \quad (P \in \{\text{ISUB, IMLTEV}\}),
\]
and for sublimation (of evaporation of melting) snow aggregates (as above for ice crystals, bar the different subscripts),

\[
\begin{align*}
R_{N_{cfa}}^{(i)} \big|_P &= \frac{\Gamma (\alpha_{cfa} + d_{cfa} + 1) T_{q_{cfa} \rightarrow q}|_P}{\Gamma (\alpha_{cfa} + b_{cfa} + d_{cfa} + 1)} \times \left( \frac{\Gamma (\alpha_{cfa} + b_{cfa} + d_{cfa} + 1)}{F_{q_{cfa}}} \right) n_{a,cfa} c_{cfa} \left( \frac{\rho_0}{\rho} \right)^{1 + \alpha_{cfa} + b_{cfa} + d_{cfa} - n_{0,cfa}} \\
R_{q_{cfa}}^{(i)} \big|_P &= \frac{m_{cfa}^{(i)}}{q_{cfa}} T_{N_{cfa} \rightarrow q}|_P
\end{align*}
\]

\((P \in \{SSUB, SMLTEV\})\). (3.122)
Chapter 4

Future plans

4.1 Outline

September 2010

- Finalise the scavenging scheme developed in Chapter 3 and discuss with other members of the UKCA project.

- Begin implementing the aerosol element of this scheme in UKCA, possibly taking advantage of either the UM single-column mode or the Kinematic Driver model\(^1\) to provide a testing environment without the complexity of the global model.

- Present a poster at the AeroCom workshop, and a talk at the AOPP retreat (both end of September) describing the cloud-cycling results of Chapter 2 as well as any preliminary results obtained from the new scheme.

Michaelmas Term 2010

- Finish implementing the aerosol element of the new scavenging scheme, with hooks to enable code sharing with chemistry and convective scavenging.

- Plan an evaluation strategy for the new scheme, to test whether it improves the comparison between aerosol parameters derived from the model and those obtained from in-situ and remote-sensing observations, with particular regard to changes in long-range transport.

- Begin multi-year simulations for the control model and each configuration of the new scheme to provide the model data for evaluation.

- Collect relevant observational data sets for evaluating the model.

- Write code (or identify/adapt existing code) to carry out the model-observation comparison.

Hilary Term 2011

- Run the evaluation and write up the results.

- Start working on the extra diagnostics required in the convection scheme to extend the cloud-cycling analysis to include cycling in convective cloud, and the new scavenging scheme to include convective scavenging properly integrated with convective transport.

\(^1\)http://www.convection.info/microphysics
4.2. IN DETAIL

Trinity Term and Summer 2011

- Re-run the cloud-cycling analysis with convective cloud included, and compare to Pruppacher and Jaenicke (1995) and Croft et al. (2010a).

- Couple the new scavenging scheme with the convection scheme to give a detailed representation of convective scavenging and re-run the evaluation, this time with particular emphasis on changes in vertical transport due to convective scavenging.

Academic Year 2011-2012

- Investigate the effect of including the more detailed scavenging representation on the strength of the various aerosol indirect effects discussed in Section 1.1.3 in the model.

- Write up thesis.

4.2 In detail

4.2.1 Questions to be answered

Once we have a detailed scavenging scheme implemented, there are a number of scientific questions we can start to address. By coupling scavenging and precipitation microphysics in a physically-detailed way, a number of parameters which are often subject to arbitrary tuning (prescribed scavenging ratios) are removed. How does the global distribution and transport predicted by such a scheme compare against that which can be achieved by careful tuning in a simpler parameterisation (as in e.g. Bourgeois and Bey, 2010)?

Aircraft campaigns have compiled detailed size-resolved vertical profiles of aerosol in limited regions, which are difficult to observe from ground- or satellite-based instruments. For example, the three HIPPO campaigns have compiled many vertical profiles along a pole-to-pole track over the Pacific. These include both size-resolved particle counts from an ultra-high sensitivity aerosol spectrometer (UHSAS) and measurements of the black carbon content of individual particles from a single-particle soot photometer (SP2); the latter are presented in Schwarz et al. (2010). This provides an important set of observations against which we can compare model-derived vertical profiles. Given the potential impact of scavenging processes on vertical transport, it will be interesting to evaluate the extent to which a detailed scavenging scheme helps the model to reproduce these observed profiles.

Finally, scavenging followed by evaporative release provides a mechanism for an upward shift of the aerosol size distribution. Given the strong size dependence of both the direct radiative effect and indirect effects via interaction with subsequently-formed cloud, cloud cycling may thus cause significant modification of both effects; with a detailed representation of this process in the model we can attempt to quantify its role.

4.2.2 Convective cycling and scavenging

In order to diagnose the cycling of water through convective clouds, we need to add diagnostics for the condensation and evaporation of cloud water within the convective plume. (Although these processes are calculated within the convection scheme, they are not presently available as diagnostics.)

To diagnose the cycling of aerosol through convective clouds, we need a way to calculate the take-up and release of aerosol by convective cloud water. The approach used for large-scale
clouds relies on the grid-box cloud fraction; however this is poorly defined in a mass-flux convection scheme such as that used in the UM. In this context, it may be preferable to use the rates of entrainment and detrainment of aerosol into the updraught and downdraught (calculated in order to perform convective transport, although not presently available as diagnostics). Entrained aerosol is then considered to all be taken up according to the scavenging ratio for a given mode, unless the updraught/downdraught is dry (contains no condensate, e.g. below the lifting condensation level), while non-entrained aerosol also takes into account the grid-box large-scale cloud fraction as described in Section 2.2.

As mentioned briefly in Section 3.1, the current approach of operator-splitting convective transport and scavenging leads to problems, as aerosol is transported all the way up the convective plume before it is seen by the scavenging scheme; only aerosol which is already present or detrained at the level where convective precipitation is formed is available for removal by convective scavenging.

This is clearly undesirable: convective scavenging should remove aerosol that is being transported through the convective plume in between entrainment and detrainment, even if neither occurs in the precipitating layer. In order to make this possible, our intention is to add “hooks” such that the scavenging scheme can be called on the in-plume quantities during convective transport. This will reduce the structural separation between the meteorological and chemistry/aerosol schemes, but this appears necessary if scavenging by sub-grid-scale convection is to be treated correctly.
Appendix A

Transferable skills

I have developed and built on a number of skills during the course of my first year, including:

- defining a problem and developing a method to analyse it (e.g. Chapter 2);
- developing a detailed model based on a conceptual framework (e.g. Chapter 3);
- finding, reading and understanding the relevance of primary scientific literature;
- developing an awareness of the broader field in which my work fits (e.g. through reading and attending seminars), and understanding where my own work fits into this (e.g. comparisons with earlier results in Section 2.3);
- networking and collaboration with others working on related projects (e.g. UM Users’ workshop at Met Office, UKCA Developers’ Meeting in Cambridge);
- delivering a short presentation at the UKCA Developers’ Meeting;
- working with a previously-unfamiliar programming language (FORTRAN);
- working in a massively-parallel high performance computing (HPC) environment, including a one-day training course at the Computing Laboratory.
- revising and extending my \LaTeX{} knowledge in the course of preparing this and other documents;
- preparing and submitting a paper based on the work for my MSc dissertation (co-authored with my two supervisors on that project, and currently awaiting peer review).


Andreae, M. O., D. A. Hegg, and U. Baltensperger, Sources and nature of atmospheric aerosols, in *Levin and Cotton* (2008), chap. 3.


Bourgeois, Q., and I. Bey, Sensitivity of aerosol transport towards the Arctic to scavenging, Presentation at ECHAM5-HAMMOZ meeting, ETH Zurich, Switzerland, 24–26 March, 2010.


Denman, K. L., et al., Couplings between changes in the climate system and biogeochemistry, in Solomon et al. (2007), chap. 7.


Forster, P., et al., Changes in atmospheric constituents and in radiative forcing, in Solomon et al. (2007), chap. 2.


