### UKCA\_ACTIVATE

Aerosol activation and indirect effects in UKCA (UM 8.4)

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### Introduction

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• From a bulk perspective, water vapour condenses to form cloud when the local relative humidity (RH) reaches 100%.



Figure : (P. Stier) Condensation as a rising parcel cools adiabatically to its dew point.



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• However, the optical and microphysical properties of the cloud depend strongly on the droplet size distribution, not just the mass of liquid water.

• The droplet size distribution, in turn, is dependent on the number of aerosol particles able to act as *cloud condensation nuclei* (CCN).



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Figure : Aerosol-induced changes in the droplet size distribution can affect both the albedo and lifetime of the cloud, affecting the radiation balance. (Forster et al., 2007)



## A brief review of Köhler theory

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Figure : (P. Stier) The saturation vapour pressure  $e_s(\infty)$ , corresponding to 100% RH, describes the point at which vapour is in equilibrium with a *plane surface* of liquid.



## A brief review of Köhler theory

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- Homogeneous nucleation of droplets can only occur at supersaturations (S = RH 1) much greater than those found in the atmosphere (typically less than 1%).
- Instead, cloud droplets form by condensation onto hygroscopic aerosol particles which act as *cloud condensation nuclei*.
- This process is described by Köhler theory, involving the competing effects of a curved surface and solute within the droplet.



## The Kelvin (curvature) effect

• Two ways of looking at this:

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- To grow a small droplet, work must be done against its surface tension. This makes smaller droplets harder to grow.
- Surface molecules feel attractive forces from fewer neighbours in a smaller droplet, so can escape more easily.



Figure : P. Stier



## The Kelvin (curvature) effect

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• Result: equilibrium vapour pressure over a convex surface is greater than over a plane surface.

$$e_{\rm s}(r) = e_{\rm s}(\infty) \exp\left(\frac{2\sigma}{rR_{\rm v}\rho T}\right)$$



## The Raoult (solute) effect

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- The Kelvin effect can be overcome by the presence of a solute (e.g. soluble aerosol such as sulphate or sea-salt).
- Result: the equilibrium vapour pressure of a solution is reduced proportionately to the mole fraction of solute.



Figure : Solute molecules reduce the number of solvent (water) molecules which are at the surface and so able to escape. (P. Stier)



## The Raoult (solute) effect

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• For a fixed mass *m* of solute in the droplet, the solution becomes more dilute as the droplet grows. For weak solutions,

$$\frac{e_{\rm s}^{\rm (sol^{\underline{n}})}}{e_{\rm s}^{\rm (pure)}} = 1 - \frac{3im\mu_{\rm l}}{4\pi r^3 \rho_{\rm l} \mu_{sol\underline{n}}} \tag{1}$$

(where *i* is the van't Hoff factor accounting for ionic dissociation).

 This allows aerosol particles containing soluble material (e.g. (NH<sub>4</sub>)<sub>2</sub> SO<sub>4</sub> or NaCl) to grow into cloud droplets at low supersaturations, overcoming the Kelvin effect.



### The Köhler equation

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• These two competing effects combine to give the supersaturation *S* at which a droplet or radius *r*, in which an aerosol particle of dry radius *a* is dissolved, is in equilibrium:

curvature parameter (depends on temperature)



hygroscopicity parameter (depends on composition)



### The Köhler curve



droplet is in upon the a

Figure : The Köhler curve shows how the supersaturation at which a droplet is in equilibrium varies according to its size. Its shape depends upon the quantity and composition of solute in the droplet. If the critical supersaturation  $S_c$  is reached, the droplet can grow without limit and is said to be *activated*. (McFiggans et al., 2006)

wet droplet radius, r.um

100

Kelvin term Raoult term Total

10

10<sup>2</sup>



### The supersaturation balance

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- The key to calculating which aerosol particles activate is thus to find the maximum supersaturation  $S_{\max}$  reached as the cloud forms.
- This is a balance between adiabatic cooling as the air parcel rises and condensation onto existing droplets:



• Setting  $\frac{dS}{dt} = 0$  leads to a nonlinear integral equation for  $S_{max}$  which cannot be solved analytically, and hence a variety of parametrisations have been developed.



### Parametrisation

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• The most detailed approaches use an explicit parcel model, numerically simulating the evolution of supersaturation and droplet size as an air parcel rises through cloud base.

This is very computationally expensive, and so schemes such as Nenes and Seinfeld (2003) and Barahona et al. (2010) instead use population splitting, limits and iterative numerical methods to solve for S<sub>max</sub> approximately.



## Parametrisation: Abdul-Razzak and Ghan

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- The parametrisation in UKCA (Abdul-Razzak and Ghan, 2000) instead uses an empirical fit to detailed numerical simulations with a parcel model to estimate  $S_{max}$ . While not as accurate as the above schemes, it is much cheaper to run.
- This is formulated for multiple internally-mixed log-normal modes, as used in GLOMAP-mode, with all modes in competition for the available water vapour.
- An implementation of Barahona et al. (2010) also exists on a branch by Daniel Partridge (now at Stockholm University).



## Parametrisation: Abdul-Razzak and Ghan

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• Once S<sub>max</sub> is known, the Köhler equation can be used to find the minimum dry radius above which aerosol particles in each mode will activate.

• It is then a simple matter of integrating the log-normal distributions to calculate the number of droplets formed.



### Dependence on vertical velocity

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- The supersaturation balance depends strongly on the vertical velocity *w*, which controls the source term.
- Within the turbulent environment at cloud base, the local vertical velocity varies considerably.
- Following West et al. (2014), the activation scheme is integrated over a 20-bin normal distribution of vertical velocity, truncated at zero.

in practice, this limit  
is hit too often...  
$$W \sim N\left(w_{\text{large-scale}}, \max\left(\frac{2}{3} \underbrace{\mathsf{TKE}}_{\text{form boundary layer scheme}}, (0.1 \text{ m s}^{-1})^2\right)\right), \quad W > 0$$
turbulent kinetic energy  
from boundary layer scheme  
– this formulation assumes isotropic turbulence



## Implementation in UKCA (West, 2012)

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- The UM has a bulk cloud scheme with no prognostic cloud droplet number concentration (CDNC).
- At (the end of) each timestep, CDNC is diagnosed as the number concentration of activated droplets calculated by the activation scheme (with a hard minimum of 5 cm<sup>-3</sup>).
- The CDNC diagnosed at cloud base is used as a uniform profile throughout any contiguous layers of liquid cloud above, as the bulk cloud scheme handles neither transport nor depletion by coalescence.



### Aerosol activation

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- Implemented in UKCA\_ACTIVATE, which is called after the rest of GLOMAP-mode, directly from UKCA\_MAIN1.
- Inputs are thus 3D (UM-style) rather than 1D (GLOMAP-style) arrays, and aerosol inputs are in tracer form (mass and number mixing ratios) rather than ND and MD arrays.
- Dry radius is obtained by pulling it back out of the mode\_diags array.
- Due to shared heritage with other implementations, the arrays are internally reshaped to 2D.



# Aerosol activation (2)

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- UKCA\_ABDULRAZZAK\_GHAN is called to evaluate the actual parametrisation:
  - calculation of curvature and hygroscopicity parameters;
  - empirical fit to calculate max. supersaturation reached;
  - calculation of how many particles activate in each mode as a result;
  - all integrated over updraught PDF.
- The diagnosed CDNC is then output to D1 and section 34 via a reserved slot in the chem\_diags array, for coupling to the radiation and precipitation schemes.
- Other diagnostics are output to section 38 via the mode\_diags array, in a set of reserved elements left untouched by UKCA\_AERO\_CTL.



### Indirect effects

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• The indirect effects work via the CDNC dependence of processes in the radiation and precipitation schemes.

• The CDNC calculated in UKCA\_ACTIVATE is passed through to these schemes via D1, much as for UKCA\_RADAER, using the interface in UKCA\_CDNC\_MOD.



# First indirect (albedo) effect: radiation coupling

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- If this is enabled in the UMUI (L\_UKCA\_AIE1=.TRUE.), the CDNC from UKCA is passed down into the radiation scheme and used in the calculation of droplet effective radius in R2\_RE\_MRF\_UMIST.
- There are many layers of forwarding routines, but it's passed through from ATMOS\_PHYSICS1 as n\_drop\_pot!
- This causes the effective radius, and hence the cloud albedo, to depend on the number of activated droplets, giving rise to the first indirect effect.



# Second indirect (lifetime) effect: precip. coupling

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- If this is enabled in the UMUI (L\_UKCA\_AIE2=.TRUE.), the CDNC is passed down into the precipitation scheme (LS\_PPN) and used in the calculation of the droplet size distribution (LSP\_TAPER\_NDROP) used in the autoconversion parametrisation (LSP\_AUTOC).
- This causes the autoconversion rate to respond to changes in the number of activated droplets, giving rise to the second indirect effect.
- The details of the CDNC dependence can be found in UMDP 26, which decribes the large-scale precipitation scheme in great detail.



## Configuration via UMUI: activation

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- In UKCA (Section 34) "MODE" sub-window, setting logicals in UKCA\_OPTION\_MOD:
  - Switch to use Abdul-Razzak and Ghan in place of the older empirical aerosol–CDNC relation. This sets L\_UKCA\_ARG\_ACT.
  - Switch to calculate diagnostics of the CCN that would activate at a range of fixed supersaturations (hard-coded in UKCA\_ACTIVATE.F90). This sets L\_UKCA\_SFIX.



## Configuration via UMUI: indirect effects

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- In UKCA "coupling" sub-window, setting logicals in UM\_INPUT\_CONTROL\_MOD:
  - Switch for the first indirect (albedo) effect in the radiation scheme. This sets L\_UKCA\_AIE1.
  - Switch for the second indirect (lifetime) effect in the precipitation scheme. This sets L\_UKCA\_AIE2.
- If these are set to .FALSE., fixed values of CDNC over land and ocean are used instead.



## Diagnostics: section 34, in D1

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Implementation	ltem	Description	Units
Aerosol activation Indirect effects	162	mean CDNC in cloudy portion of grid box	$m^{-3}$
First indirect (albedo) effect	163	mean $CDNC^{-1/3}$ in cloudy portion of grid box	m
Second indirect (lifetime) effect	100		



## Diagnostics: section 38

CA_ACIIVAIE	ltem	Description	Units
Zak Kipling	469	Mean no. conc. of activated NUCSOL in cloudy portion	$m^{-3}$
	470	Mean no. conc. of activated AITSOL in cloudy portion	$m^{-3}$
	471	Mean no. conc. of activated ACCSOL in cloudy portion	$m^{-3}$
	472	Mean no. conc. of activated CORSOL in cloudy portion	$m^{-3}$
	473	$S_{\max}$ : max. supersaturation in strongest updraught bin	%
	474	Cloud base flag (1 at base, 0 elsewhere)	1
	475	$\sigma_w$ : st. dev. of updraught PDF	${ m m~s^{-1}}$
	476	Liquid cloud fraction as seen by activation scheme	1
	477	Grid-box mean $CDNC = (34, 162) \times (38, 476)$	$m^{-3}$
	478	Liquid cloud flag (1 if liquid present, 0 elsewhere)	1
onfiguration iagnostics	479	Cloud-masked CDNC = $(34, 162) \times (38, 478)$	$m^{-3}$
	480	Cloud-masked $S_{max} = (38, 473) \times (38, 478)$	%
	481	Cloud-masked $W_{char}$ (uniform updraught which would	${ m m~s^{-1}}$
		activate the same number of droplets as the full PDF)	
	482	Cloud-masked $\sigma_{w} = (38,475)  imes (38,478)$	${ m m~s^{-1}}$
	483	Cloud-masked TKE as seen by activation scheme	$m^2 s^{-2}$
	484	Surface CCN at fixed supersaturation (each level in diag.	$m^{-3}$
		represents a different supersaturation) only if enabled	



## Evaluation: aircraft campaigns





Figure : CDNC simulated by UKCA (in UM 7.3, using both fixed and TKE-dependent  $\sigma_w$ ) compared to those observed during various aircraft campaigns. (West et al., 2014)



## The global picture

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Figure : Simulated CDNC and total anthropogenic radiative flux perturbation (RFP).



### Vertical velocity issues

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- The TKE-based vertical velocity hits the lower limit too often, which is not ideal. There are several possible reasons for this:
- Cloud base occuring outside the region where the boundary-layer scheme calculates TKE.
- Condensation due to updraughts parametrised by the convection scheme rather than turbulence or large-scale ascent.
- The mismatch between prognostic condensate and diagnostic droplet number.



## Finally, a note on forcing

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- The first indirect (albedo) effect can be studied using the instantaneous radiative forcing diagnostics in a "double-call radiation" run, just like the direct effect.
- This is not true for the second indirect (lifetime) effect, which only modifies the radiation budget via feedbacks from the hydrological cycle.
- Studies including the second indirect effect must use "single-call radiation" simulations in which the aerosol feedbacks are active.



### References

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