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Key outcomes/non technical summary

Current progress in the development of the UKCA model for HadGEM3-ES is reviewed against the requirements outlined by O'Connor (2011). The development of UKCA has been aided by the MONSooN joint computor resource which allows access from all the UKCA developers. Recent developments to UKCA are listed below.

Chemical schemes designed to address both tropospheric and stratospheric chemistry, and capable of oxidising Biogenic Volatile Organic Carbons have now been extensively tested in UKCA.

Chemical schemes for aerosol production in the troposphere and stratosphere are also available and can be added to any other chemical scheme. A tropospheric heterogenous chemistry has been tested.

The UKCA-MODE aerosol scheme is an established component of UKCA and recent work has included the addition of mineral dust aerosol, stratospheric aerosol, and boundary-layer nucleation process.

A new scheme for indirect forcing from aerosols is available which uses the results from the UKCA-MODE aerosol scheme.

The capability of the HadGEM3A model to simulate the dynamical features which are essential for correct operation of the stratospheric chemistry is being studied. The effects of tuning the non-orographic gravity wave drag scheme are shown.

Recent improvements and ongoing work are making the UKCA model more self- consistent so that, for example, changes to aerosol concentrations change the chemistry through the photolysis rates. The improvements will also ensure that the model responds to changes in clouds and precipitation through wet deposition processes and the photolysis scheme, and thus ensure that the response to climate change can be modelled.

Associated publications

Press interest

None.

Progress Report on UKCA Development for HadGEM3

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Contents

1 Introduction

The United Kingdom Chemistry and Aerosol (UKCA) model is a community model being developed through a joint programme involving scientists working at the universities of Cambridge, Leeds, and Oxford, and at the Met Office. Work on the development of UKCA is also being carried out in New Zealand (NILU), and Australia (CSIRO). Although this report is concerned on the application of UKCA in the global climate model, UKCA is also being used for air quality prediction in a limited area forecast model, and is also being ported to the regional climate model.

The UKCA model has been developed to provide a suitable model environment for coupled aerosol-chemistry modelling with the Unified Model. It contains code for the prediction of important gas-phase and aerosol species in the atmosphere, in particular for those important in the radiative forcing of the global climate. UKCA remains a flexible model, with both aerosol and chemistry components being configured through the Unified Model User Interface. Mann et al. (2010) describes the aerosol scheme that has been developed for UKCA, while Morgenstern et al., (2009) describes the stratospheric ozone model.

In the HadGEM2 Earth System Model, a description of tropospheric chemistry was used in order to prediction of tropospheric ozone and methane together with the tropospheric oxidants used in the generation of sulphate and nitrate aerosol. Here we explore recent improvements made to the UKCA model, with a focus on the work which is likely to contribute to the objectives of the HadGEM3 Earth System Model (HadGEM3-ES).

O'Connor (2011) reports on the objectives of HadGEM3-ES and discusses the potential improvements to Earth-System modelling which could be made with UKCA, and the following topics concerning UKCA are relevant here:

1) The chemical scheme should encompass both stratospheric and tropospheric chemistry, in order to provide a seamless ozone prediction.
2) Methane concentrations should be predicted from natural and anthropogenic emissions.
3) Interactions between the biosphere and chemistry/aerosol modelling should be enhanced, implying that more sophisticated chemical schemes are required for biogenic volatile organic carbons.
4) An online photolysis scheme should be used so that the photolysis rates are calculated for current timestep conditions and reflect for example, the ozone column and the atmospheric aerosols which are simulated in UKCA.
5) The UKCA model is expected to become more self-consistent, with products available from different modules being used in different parts of UKCA. For example, the results from the UKCA-MODE model will be used as to modify the predicted photolysis and as an input for the heterogenous chemistry routine, as well as being used to provide radiative forcing.
6) The UKCA-MODE aerosol scheme should be used instead of the CLASSIC model, as this is likely to improve the radiative forcing estimates.
7) An improved indirect radiative forcing scheme should be used.

Here we present a description of recent improvements to UKCA, and how these relate to the development of the HadGEM3-ES model.

2 Chemical Schemes

A variety of chemical schemes have been developed for UKCA, and those with the potential for HadGEM3-ES development work are:

1) Stratospheric Chemistry [Strat],
2) Tropospheric Chemistry: The MIM isoprene scheme [MIM],

3) Tropospheric Chemistry: The Extended tropospheric scheme [ExtTC]

3) Chemistry for Troposphere and Stratosphere [CheST]

Each of these chemistries is described below and have an optional aerosol scheme which is suitable for the production of aerosol from gas phase species in the troposphere and stratosphere as required. The chemical schemes are now all integrated by the Newton-Raphson chemical solver.

2.1 Stratospheric Chemistry (Strat)

Stratospheric chemistry in UKCA covers the oxidation of CH4 and CO and the interaction of the halogens Cl and Br with the cycles of HOx, NOx and Ox. In total there are 37 chemical tracers undergoing 169 photochemical reactions. The current mechanism is described in detail in Morgenstern et al. [2009] and has been used extensively in theCCMVal project [Eyring et al [2010], Morgenstern et al. [2010a,b]]. Testing of this model at a resolution of N48L60 in a coupled atmosphere-ocean model is ongoing, and (figure 1) shows the total column ozone from the model. Comparison of this with TOMS/SBUV data shows that this reproduces all the major features, with a well developed ozone hole over the Antarctic.

2.2 Tropospheric Chemistry (MIM)

The tropospheric chemistry scheme in UKCA details the oxidation of methane, ethane, propane, and acetone as described in Law et al., [1998] with updates to the kinetics following IUPAC recommendations (2006). Isoprene oxidation is also included with the present scheme based on the Mainz Isoprene Mechanism (Pöschl et al., 2000). In this scheme, there are 60 chemical tracers participating in 132 photochemical reactions. A description of the tropospheric chemistry scheme is given in Telford et al. (2010). Archibald et al. (2011) compare the impacts of modifications to the isoprene oxidation mechanism and show that the current tropospheric chemistry scheme compares well against ozone sonde observations as well as the CMDL flask sampling network for carbon monoxide.

2.3 Extended Tropospheric Chemistry (ExtTC)

The extended tropospheric chemistry scheme includes descriptions for the photochemistry of methanol, ethene, ethyne (acetylene), propene, butane, a lumped aromatic hydrocarbon and alpha-pinene in addition to the species and reactions covered by the tropopsheric chemistry scheme. A description of the scheme is given by Young (2008). The extended scheme increases the total number of tracers by 20 and the total number of reactions by 80 compared with the tropospheric chemistry scheme.

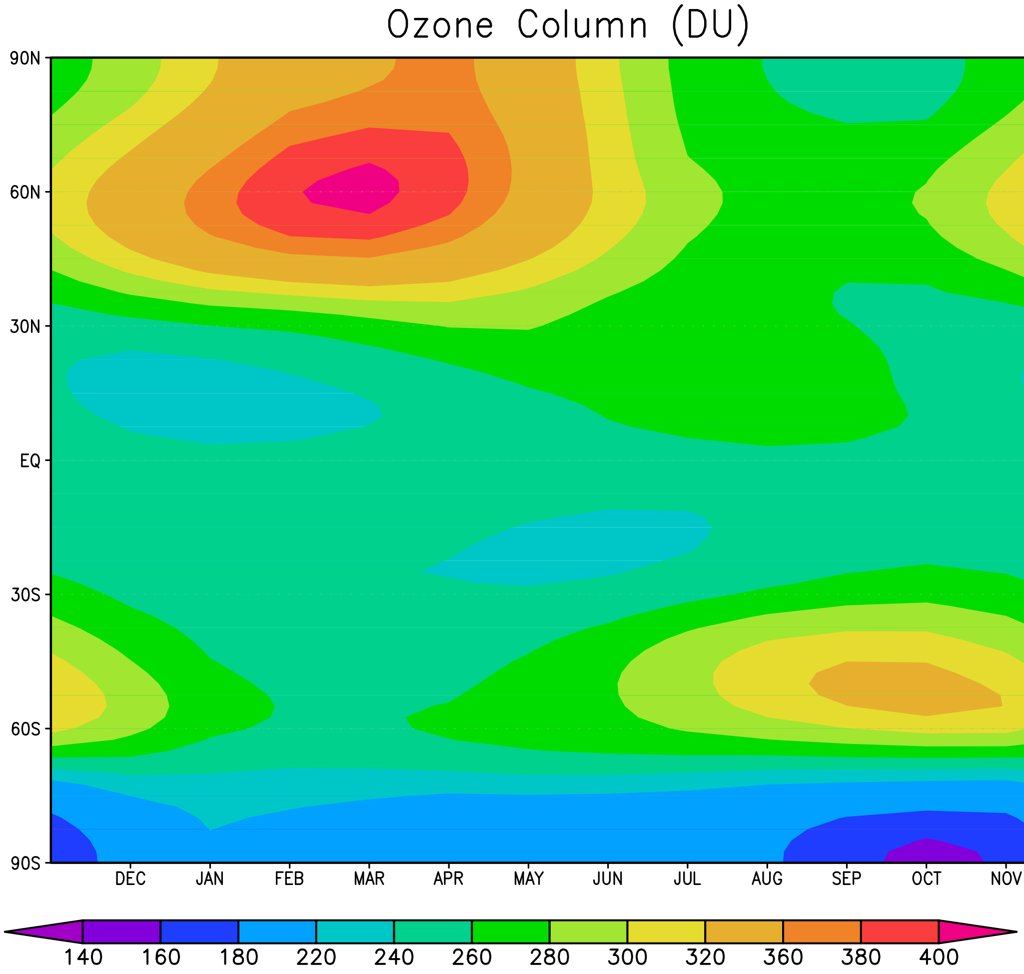

Figure 1: Mean ozone column (DU) from 21-year model simulation using Stratospheric Chemistry of a 1990 state using a coupled atmosphere-ocean model at N48L60 resolution.

2.4 Chemistry for Troposphere and Stratosphere (UKCA-CheST)

A merged chemistry scheme has been created by combining the species and reactions from the stratospheric chemistry with those used in the tropospheric chemistry scheme. Where appropriate duplicate reactions have been removed and in specific cases where kinetic data differed between duplicate reactions the tropospheric chemistry scheme kinetics were chosen. This scheme provides a holistic basis for performing long integrations to look at the coupled effects of chemistry and climate. In total there are 75 species that undergo 281 reactions.

To support the development of UKCA-CheST a comprehensive comparison of model results with tropical and subtropical ozone sonde stations from the SHADOZ data base has been performed (http://croc.gsfc.nasa.gov/shadoz). The high resolution SHADOZ sonde data is binned in a way as to simulate the models vertical resolution, and the model issampled at the stations location. Figure 2 shows an example for Alajuela in Costa Rica (84.21W, 9.98N). The station climatology is derived from soundings during the years 2007-2009. The annual mean ozone partial pressure profiles (left panel) from Alajuela (red) and a recent UMUKCA-CheST model run (blue) are in good agreement. The rapid ozone increase above 100hPa is well captured, indicating a well modelled tropical tropopause. The annual cycle of ozone partial pressure as a function of ambient pressure (middle panel for UMUKCA-CheST, right panel for Alajuela) is well represented in the model. The timing and amplitude of the tropospheric spring and stratospheric autumn maximum is well captured. Small differences are visible during autumnin the free troposphere, with the model having lower ozone than observations. Overall, the agreement between model and observations is reassuringly good.

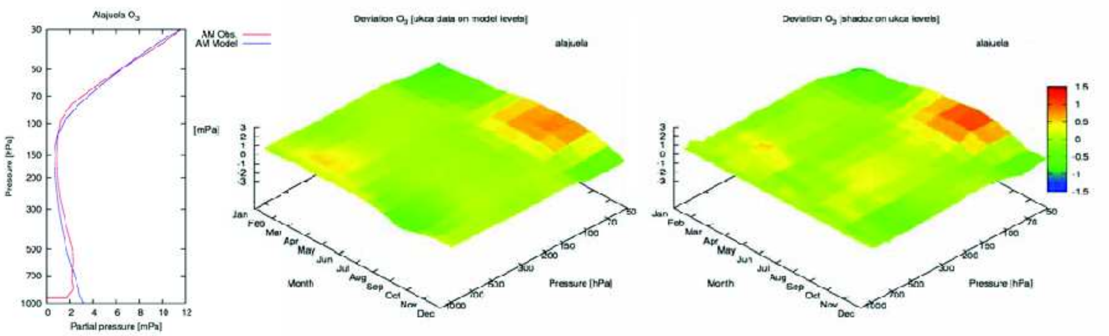

Figure 2: Ozone partial pressure for Alajuela and UMUKCA-CheST. Left panel: annualmean; middle panel: mean annual cycle for UMUKCA-CheST; right panel: mean annual cycle for Alajuela.

2.5 Comparison of tropospheric model results with satellite measurements

A range of results from satellites is available to compare with model results. Morgenstern et al., (2009) show the comparison of simulated ozone from the UKCA stratospheric chemistry model with results from a Haloe/MLS climatology, and also shows comparisons for NO_x, N_2O, HCl, and other species. For the tropospheric model, Voulgarakis et al. (2011) show the comparison between tropospheric ozone and carbon monoxide from UKCA model results and the Tropospheric Emission Spectrometer (TES) instrument aboard the NASA Aura satellite. This paper also analyses the correlation between model and measured O_3 and CO correlation. The spatial distribution of O_3 measured by TES is reproduced quite well in UKCA, however UKCA results look somewhat larger than the measurements, however TES ozone is thought to be overestimated.

3 Chemical Solver

The chemistry schemes described above are built within the framework of the ASAD chemical integration package (Carver et al., (1997)), which includes solving the chemical reactions together with wet and dry deposition. Using ASAD is a flexible approach that allows for the easy addition of species, tracers, and reactions.

ASAD can use a number of different chemical solvers, but currently UKCA uses a symbolic sparse-matrix Newton-Raphson solver (derived from Wild and Prather, (2000)) with a 1-hour timestep. This solver ensures convergence at each grid-point every timestep and is robust in changes to vertical and horizontal resolution or model top. Recent work with the solver includes adding the capability for product yields to be taken into account so that more complex chemistries can be solved. Work is proceeding on the optimisation of the Newton-Raphson solver for the IBM computor.

4 Photolysis Schemes

An online, interactive, photolysis scheme is being implemented within UKCA to allow photolysis rates to be produced that reflect the current distribution of clouds, ozone and aerosol at every timestep. This allows for feedback from the climate, (through changes in cloud optical depth) and from the aerosol model, (from changes in aerosol optical depth), to the chemistry to be included. This approach has been shown to provide a better description of the variability of trace gases such as ozone (Voulgarakis et al., 2009), thus providing better simulation of extremes such as those found in ozone episodes.

For our interactive photolysis we adopt the widely used Fast-JX scheme (Wild, 2000; Neu, 2007) which works in a consistent manner from the surface to the stratosphere. This has been implemented in UKCA and is undergoing validation (Telford et al, 2011). A plot showing its preliminary performance is shown in figure 3. It can be seen that the addition of Fast-JX greatly improves the ability of the model to simulate the seasonal cycle of ozone, especially in the southern hemisphere, for only a small reduction in speed (10-15% depending on atmospheric conditions).

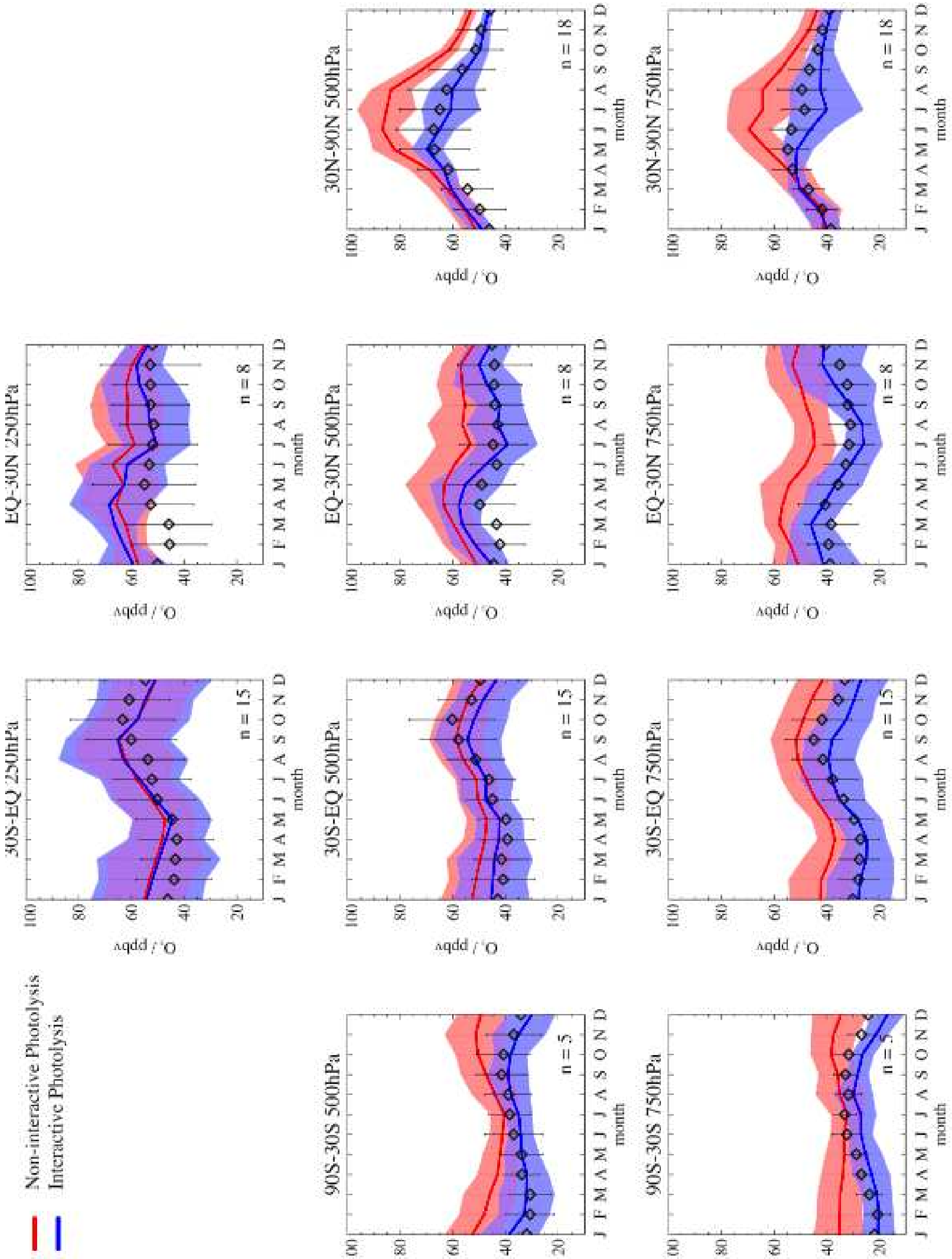

Figure 3: Comparison of O_3 concentrations between UKCA simulations, with (blue line) and without (red line) interactive photolysis, with sonde data. The comparisons are shown split into altitude and latitude bands as used by Stevenson (2006).

5 Aerosol Chemistry

5.1 Aerosol-chemistry extension for tropospheric chemistry

An aerosol chemistry describing the oxidation of sulphur dioxide and dimethylsulphide has been added and tested in a tropospheric UKCA model using the Newton-Raphson solver. In addition, a simple monoterpene oxidation scheme (Spracklen et al., 2006) is included to simulate secondary organic aerosol production. The scheme produces a secondary condensable organic species which contributes to particle growth and can also affect nucleation rates in the model. This tropospheric aerosol-chemistry scheme provides concentrations of sulphuric acid and the oxidised-organic-species to the MODE aerosol scheme for nucleation and aerosol growth by deposition (condensation), and also provides aqueous-phase oxidation rates for cloud processing of aerosol.

The tropospheric aerosol-chemistry adds only a small cost to the model, adding only seven tracers to the model.

5.2 Aerosol-chemistry extension for stratospheric chemistry

In addition to the tropospheric aerosol-chemistry scheme described above, an aerosol-chemistry suitable for the stratosphere has been added to the UKCA stratospheric UKCA model with the Newton-Raphson solver.

This aerosol-chemistry so-far just includes sulphur chemistry but includes additional species to the tropospheric sulphur chemistry resolving the sulphur tri-oxide (SO3) product and subsequent hydrolysis to sulphuric acid (H2SO4) as this intermediate step becomes important in the stratosphere. The production of SO2 in the stratosphere has a significant contribution derived from the oxidation of carbonyl sulphide (COS) and this is also included within the scheme along with photolysis reactions for COS, SO2, SO3 and H2SO4.

6 Tropospheric Heterogenous Reactions

The UKCA-MODE aerosol model and the recent transfer of the tropospheric chemistry to use the Newton-Raphson solver has created the opportunity to add heterogenous reactions on aerosol surfaces to the tropospheric chemistry in UKCA. UKCA-MODE has diagnostics (e.g. the wet surface area concentration) which should be more accurate than those of earlier aerosol schemes, making the calculation of the heterogenous rates more accurate.

Here we describe the heterogenous chemistry added to UKCA, together with some first results from a UKCA model run. The inclusion of heterogenous chemistry is important not only because of its importance for tropospheric oxidants (Jacob, 2000), but because of its impact on the generation of nitrate aerosol (Riemer, et al., 2003). McIntyre and Evans (2010) conclude that an accurate description of the uptake of N_2O_5 is required for the adequate simulation of ozone burdens and long-range transport of pollutants in the northern extra-tropics.

The reaction coefficient of the species which is adsorbed on an aerosol surface (k) is (Dentener, 1993):

$$k = (\frac{r}{D_g} + \frac{4}{\nu \times \gamma})^{-1} \times A, \tag{1}$$

where r is the aerosol radius, D_g is the gas-phase diffusion coefficient, ν the mean molecular speed, and γ is the reaction probability. A is the aerosol surface concentration in units of $[cm^2\ cm^{-3}]$, calculated as an integral over the log normal distribution using the wet radius calculated in UKCA-MODE.

Two tropospheric heterogenous reactions on aerosol are considered here:

$$N_2O_5 \Rightarrow 2\ HNO_3, \tag{2}$$

and

$$HO_2 \Rightarrow 0.5\ H_2O_2. \tag{3}$$

Both N_2O_5 and HO_2 have uptake coefficients which are estimated for aerosol surfaces composed of sulphate, black carbon, organic carbon, sea-salt, and dust. Secondary organic aerosol is assumed to behave in the same way as organic carbon. For the results presented here, dust surfaces are not included.

The reaction probabilities for N_2O_5 ($\gamma_{N_2O_5}$) are estimated using the equations outlined in Evans and Jacob (2005), although those for dust have been increased by an order of magnitude following the results of Mogili et al. (2006). For reaction on sea-salt, $\gamma_{N_2O_5} = RH \times 0.0005$ following Thornton and Abbatt (2005). The reaction probabilities for HO_2 (γ_{HO_2}) are estimated for the same surfaces using a variety of sources which have been summarised in Mao et al. (2010).

Figure 4 show the cross section of the calculated heterogenous rate coefficients for N_2O_5 for October, The calculated rate coefficient for N_2O_5 is substantially less than results from earlier models which have rates up to 5×10^4. This is due to the lower uptake coefficients used for sulphate in this study.

As expected, N_2O_5 decreases throughout the domain, and this represents a large fraction of its surface concentration. The loss of N_2O_5 represents a loss of NO_x, as reactive species are transferred to HNO_3 which is longer lived and has significant losses through wet and dry deposition. Figure 5 shows the changes to near-surface ozone, which amount to reductions of around 5 ppbv in the northern hemisphere.

The ozone concentrations predicted by the model, together with those from a control run with no heterogenous chemistry was compared with measurements taken from the ozone sonde climatology of Logan (1999). Although this data set doesn't represent the current state of the atmosphere, it has been subjected to rigerous quality checks. The distribution of stations in this dataset is biased in favour of the northern hemisphere.

Table 1 shows some statistical comparisons at different heights in the atmosphere. The comparison was done using all months, and included forty ozonesonde sites. The inclusion of heterogenous chemistry results in reductions in both the mean and root mean square deviations between the model and measurements in the lower part of the troposphere. The mean deviation was reduced by around 40 % between 1000 to 500 hPa, with the RMS deviation reduced by about 18 %. The inclusion of heterogenous chemistry did not

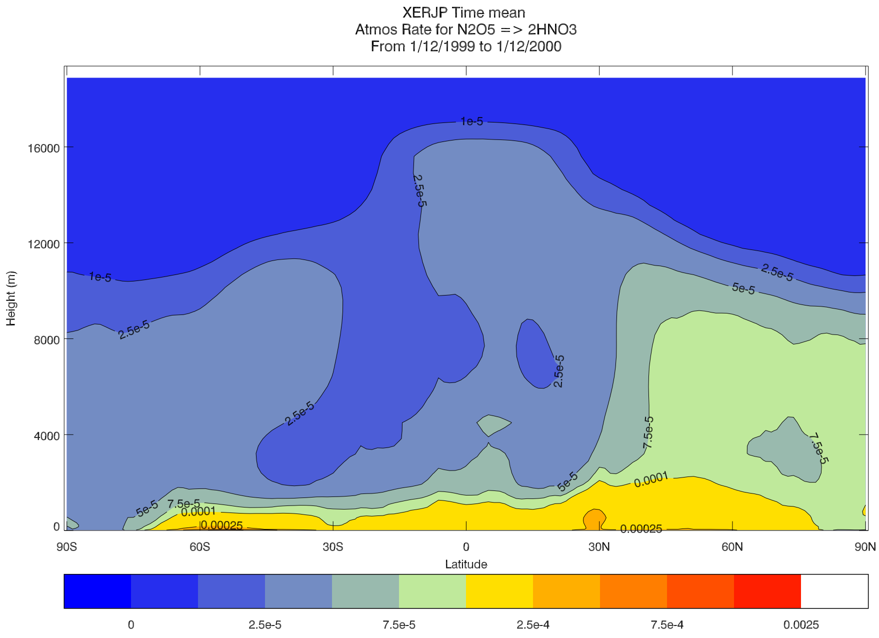

Figure 4: Zonal and annual mean heterogenous reaction rate for N_2O_5.

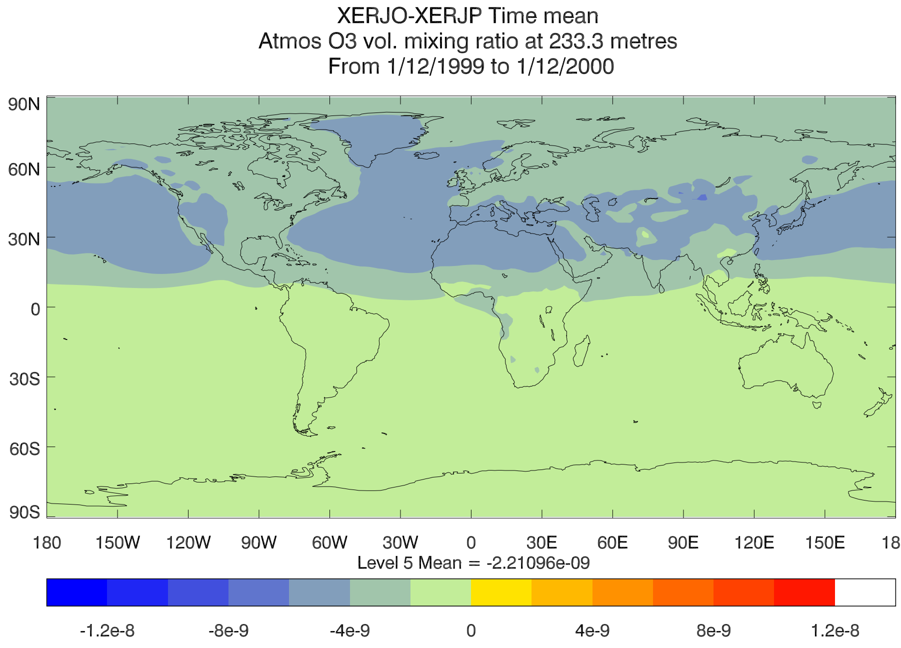

Figure 5: Change in O_3 near-surface concentrations with the inclusion of heterogenous reactions.

affect the predicted ozone concentrations in the upper troposphere above 300 hPa and lower stratosphere.

Table 1: Statistical comparison between model results and ozone sonde data from Logan (1999).

Pressure (hPa)	Correlation Cofficient	Mean Deviation (ppbv)	RMS Deviation (ppbv)
Simulation with heterogenous chemistry			
100	0.96	82.1	142.9
200	0.95	-16.6	49.2
300	0.87	2.0	18.1
500	0.70	6.7	14.2
800	0.69	6.6	12.7
900	0.66	4.7	11.0
1000	0.49	3.7	12.1
Control simulation			
100	0.96	82.2	142.9
200	0.95	-15.6	49.0
300	0.86	5.0	19.1
500	0.70	11.5	18.1
800	0.71	11.1	16.2
900	0.69	8.1	13.0
1000	0.51	6.7	13.7

7 Modifying the stratospheric zonal wind and temperature in HadGEM3

The climatology and variability of the simulated dynamical fields in a model stratosphere need to be realistic if the interactive chemistry is to generate realistic results. For example, polar stratospheric clouds (PSCs) form in the winter mid-stratosphere at temperatures below 195 K (nitric acid trihydrate clouds) or 188 K (ice clouds) (Pawson et al., 1999) so a model temperature bias here of just a few degrees will drastically alter the simulated area of PSCs and the subsequent amount of ozone depletion which occurs. The quasi-biennial oscillation (QBO) of the tropical zonal mean zonal wind is also important, not only as a significant component of dynamical tropical variability but also through the induced QBO in ozone (Butchart et al., 2003).

The parametrised non-orographic gravity wave drag scheme in the HadGEM models (Warner and McIntyre, 1999; Scaife et al., 2002) can be used to modify the extratropical stratospheric temperature and the period of the tropical QBO by changing the coefficient of the non-orographic gravity wave flux amplitude at the launch level, denoted FA_0 here. The standard model configuration sets $FA_0 = FA_{0c}$, a constant, independent of latitude. In general, increasing FA_{0c} will increase the winter extratropical stratospheric temperatures and decrease the period of the QBO. Thus there is a trade off between realistic extratropical temperatures and realistic QBO period, with the value of FA_{0c} chosen

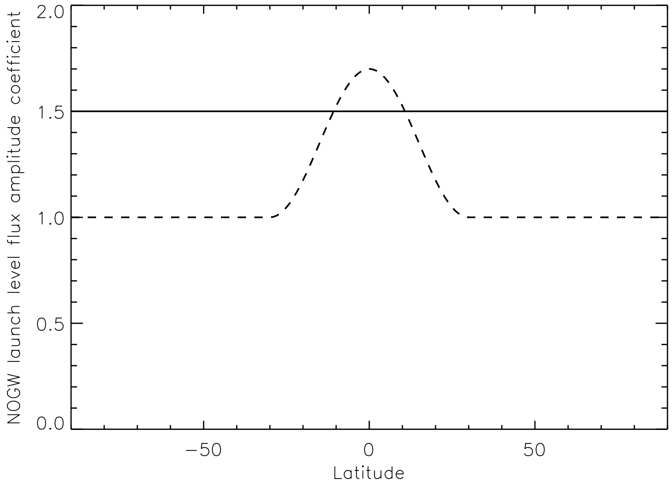

Figure 6:
Non-orographic gravity wave (NOGW) launch level flux amplitude coefficient used in constant flux amplitude (CFA; solid line) and latitudinally varying flux amplitude (LVFA; dashed line) simulations.

to give the best possible solution. However, it is likely that in reality there is a greater source of gravity waves in the tropics than in the extratropics, so it is reasonable to set

$$FA_0 = \begin{cases} FA_{0c} + FA_{0T}\ \cos^2(3\phi) & -30^\circ < \phi < 30^\circ, \\ FA_{0c} & \text{otherwise.} \end{cases} \tag{4}$$

Now the extratropical temperatures and the QBO period can be modified independently by changing the values of FA_{0c} and FA_{0T} respectively.

To investigate the effects of this change, two 10 year HadGEM3 simulations, one with constant launch level flux amplitude coefficient (CFA) and one with latitudinally varying launch level flux amplitude coefficient (LVFA) are compared. The CFA simulation uses $FA_{0c} = 1.5$ and the LVFA simulation uses $FA_{0c} = 1.0$ and $FA_{0T} = 0.7$, see Figure 6. Both simulations used a HadGEM3 atmosphere model at a resolution of N96L85 (1.875^o E-W by 1.25^o N-S, with 85 levels), with a model top of 85 km.

Figure 7 shows model temperature biases in northern hemisphere winter (December–January–February, hereafter DJF) and southern hemisphere winter (June–July–August, hereafter JJA) in both CFA and LVFA simulations. The model biases are relative to the temperatures in the UKMO stratospheric analysis (Swinbank and O'Neill, 1994). Relevant to PSC formation is the temperature at around 50hPa, averaged from 60° to 90°, in the winter hemisphere. These temperature biases in DJF and JJA in the CFA simulation are 2.73 K and 3.79 K respectively. By decreasing FA_0 in the extratropics from 1.5 to 1.0 in the LVFA simulation the biases are improved to 1.03 K and 2.29 K respectively (Figure 7).

Modifying the polar night jet strength and extratropical temperatures remains a trade off and the greater temperature biases in the winter upper stratosphere in Figure 7 (d) than those seen in Figure 7 (b) is an indication of a less realistic polar night jet strength in JJA

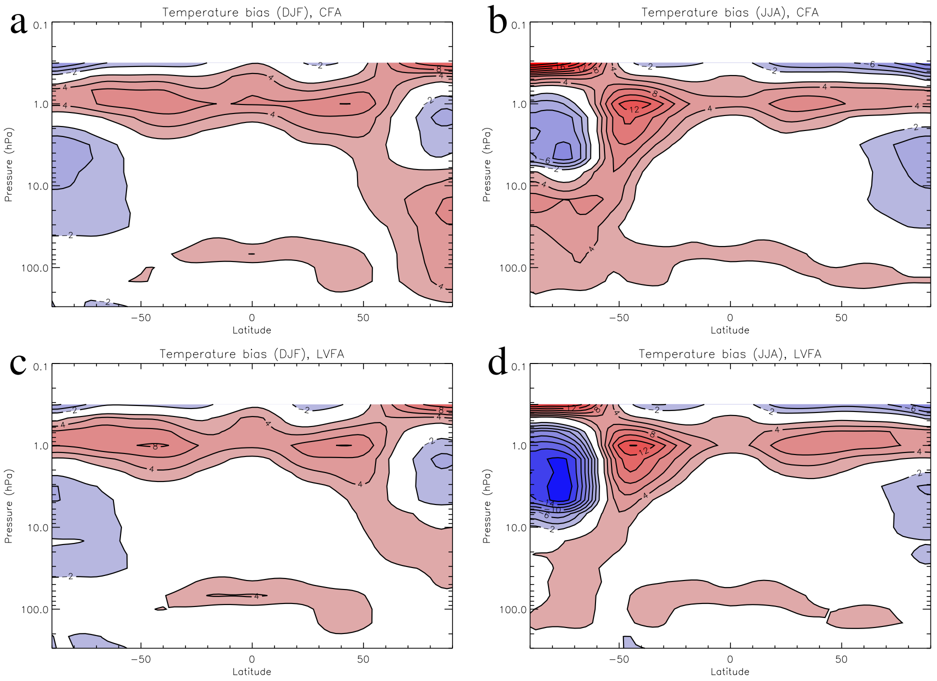

Figure 7:
Climatological temperature biases (HadGEM3 - UKMO stratospheric analysis) for (a) DJF, CFA simulation, (b) JJA, CFA simulation, (c) DJF, LVFA simulation, (d) JJA, LVFA simulation. Red/Blue shading shows a positive/negative bias. Contour interval is 2K.

in the LVFA simulation. Furthermore the tropical tropopause temperature bias of 2–4 K, although much improved in HadGEM3 over earlier model versions, cannot be modified using the non-orographic gravity wave drag scheme.

Figure 8 shows the QBO of the tropical zonal wind in both CFA and LVFA simulations. The period of the QBO in the CFA simulation is 33.5 months. By increasing FA_0 in the tropics from 1.5 to around 1.7 in the LVFA simulation this period is decreased to 25.5 months, much closer to the observed value of 27 months.

8 Developments to the UKCA-MODE aerosol scheme

8.1 Improvements to UKCA-MODE and benchmarking against GLOMAP-bin

The UKCA-MODE aerosol scheme was developed in the TOMCAT offline global chemistry transport model (CTM) and its performance in that model environment has been comprehensively evaluated (Mann et al, 2010). When used in TOMCAT, the aerosol scheme is known as GLOMAP-mode. Additionally, a paper is currently being written (Mann et al, 2011, in prep.) which benchmarks UKCA-MODE against the more detailed GLOMAP-bin scheme (Spracklen et al, 2008) in the CTM environment. During this benchmarking activity, several minor improvementns have been made (see Mann et al, 2011, in prep.) to the scheme which improves on the version in Mann et al (2010) and these have now

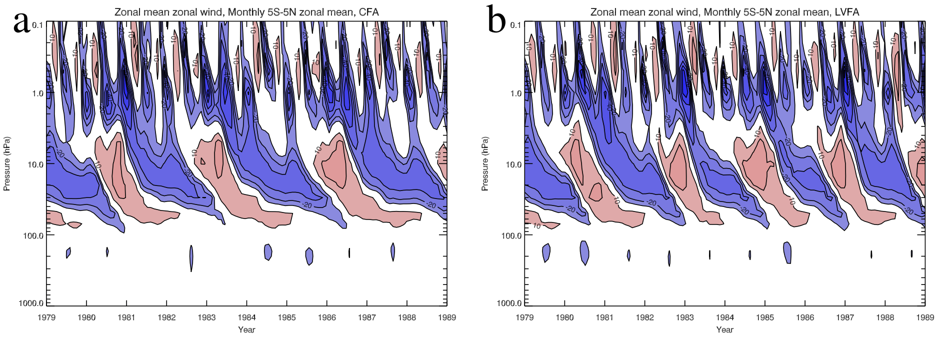

Figure 8:
QBO of the tropical zonal mean zonal wind in (a) CFA simulation, (b) LVFA simulation. Red/Blue shading shows eastward/westward wind. Contour interval is 10m s^{-1}.

been incorporated in the version maintained at the Met Office.

8.2 New parameterizations for nucleation in the boundary layer

New particle formation (aerosol nucleation) has been observed to occur globally over many different regions both within the boundary layer (BL) and in the upper free troposphere (FT) (Kulmala et al., 2004). The current nucleation mechanism implemented in UKCA-MODE (Kulmala et al, 1998) represents the FT nucleation process well (Mann et al, 2010) but does not generate nucleation events as are observed in continental regions (see Spracklen et al., 2010).

The recent updates to UKCA-MODE, as well as including general improvements to the scheme, have added new parameterizations (Metzger et al, 2010; Paasonen et al, 2010) which extend the Kulmala et al (1998) nucleation scheme to allow simulation of nucleation events in the BL. Such schemes have already been tested in GLOMAP-bin (Spracklen et al, 2010; Metzger et al, 2010) and these are included in the latest version of UKCA-MODE as well as in the main UKCA FCM branch at the Met Office.

As part of the EU FP6 EUCAARI European Aerosol Integrated Project, UKCA simulations were carried out to investigate the role these nucleation processes play in determining the properties of European aerosol. The EUCAARI project revolved around a major field campaign in May 2008 together with a new network of aerosol supersites (the EUSAAR network) were employed to make detailed measurements of aerosol size distribution and composition.

As part of a joint EUCAARI-AEROCOM model intercomparison around nucleation processes, UKCA-UM simulations were carried out with UKCA-MODE coupled to the UKCA StdTrop-Aerchem chemistry and the UM winds and temperatures nudged towards ECMWF operational re-analysis fields for the year 2008. The model was set to output the model tracers at the surface interpolated to each of the EUSAAR sites at each model timestep thus enabling high temporal resolution evaluation of the size-resolved aerosol and composition from UKCA against the detailed observations from EUCAARI.

Equivalent runs were also carried out with the GLOMAP-mode scheme driven by offline-oxidants in the TOMCAT chemistry transport model – and by parallel runs with the more sophisticated GLOMAP-bin scheme. A regional aerosol model PMCAMx-UF (Spyros Pandis) was also run in a similar fashion. Figure 9 shows this EUCAARI-AEROCOM model intercomparison of the high temporal resolution data for the concentration of particles larger than 100nm from these four size-resolved aerosol models against observations at the Hyytiala site in Finland.

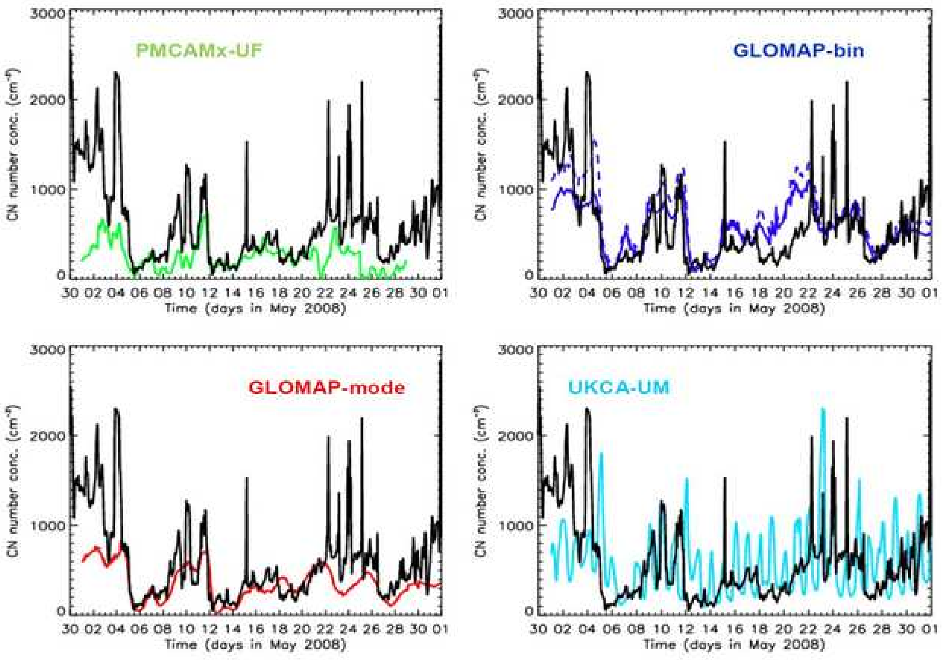

Figure 9: The number concentration of particles larger than 100nm at Hyytiala (Finland) as simulated by PMCAMx-UF (top-left), GLOMAP-bin (top-right), GLOMAP-mode (bottom-left) and UKCA-UM (bottom-right). In each case the model (interpolated to the location of the site) has been output at hourly temporal resolution and is compared to the observations which are shown by the black line.

8.3 Modifications to UKCA-MODE to simulate stratospheric aerosol

As explained earlier, a recent update to UKCA has involved an aerosol-chemistry extension to the UKCA StratChem chemistry scheme to include a sulphur chemistry scheme suitable for the stratosphere including photolytic and thermal reactions for COS, SO_2, SO_3 and H_2SO_4.

As well as adding the stratospheric sulphur chemistry, some changes to the UKCA-MODE routines were required to enable the code to applied to simulate stratospheric aerosol. In the troposphere, sulphuric acid in the aerosol phase has essentially zero vapour pressure. In the stratosphere however, the aerosol H2SO4 vapour pressure becomes sigificant and in some regions of the stratosphere, evaporation of H2SO4 can occur and can play an important role in determining the distribution of stratospheric aerosol (SPARC, 2006).

The UKCA-MODE aerosol routine for condensation has been modified and now calculates the H2SO4 vapour pressure allowing evaporation or condensation of sulphuric acid depending on the atmospheric conditions (following Ayers et al., 1980). The Kelvin effect is also included according to the mean radius calculated by GLOMAP-mode.

An option to use the nucleation scheme of Vehkamakki et al (2002) (which is applicable to both tropospheric and stratospheric conditions) has also been added, as an alternative to the Kulmala et al (1998) parameterization, which is only appropriate for the troposphere.

As a test for this StratChem+MODE configuration, the UKCA model was integrated within N48L60 HadGEM3-A with three years spin-up prior to a simulated Pinatubo eruption in June 1991 whereby 20 Tg of SO_2 is injected between the tropopause and 28 km at 15^o N, 120^o E. The model was then integrated for a further four years post-Pinatubo to investigate the evolution of the stratospheric aerosol and its size distribution.

To evaluate the UKCA simulations, we have compared to the balloon-borne profile measurements of size-resolved number concentration at Laramie, Wyoming. These measurements (Deshler et al, 2003) form an extremely good observational record against which to evaluate the model, with balloons launched at approximately monthly frequency throughout this period. Figure 10 compares model profiles of the number concentration of particles larger than 10 nm, 150 nm, and 250 nm against those from observations taken at 3 months prior to the eruption, and 9 months afterwards.

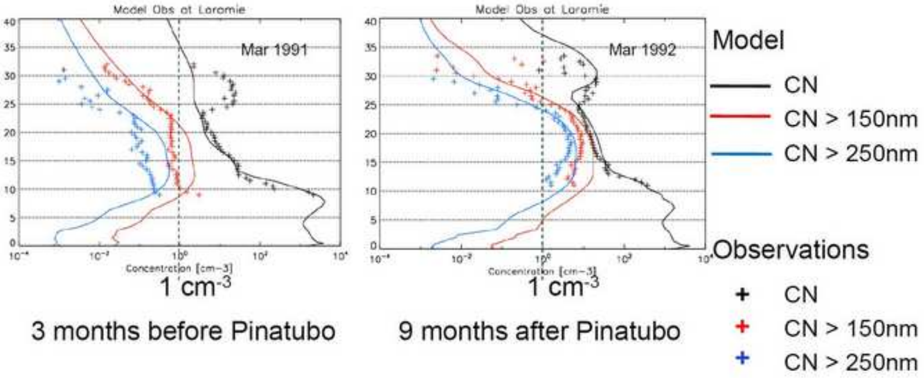

Figure 10: Comparison of profiles of size-resolved number concentrations of particles larger than 10nm (black), 150nm (red) and 250nm (blue) 3 months before, and 9 months after, the Pinatubo eruption at Laramie, Wyoming. Model profiles (N48L60 UKCA StratChem+GLOMAP) are the solid lines, observations (Deshler et al, 2003) are the asterisks.

8.4 Mineral dust now incorporated in UKCA-MODE

The UKCA-MODE aerosol model has already been developed to treat dust in the size modes used by the model (see Mann et al, 2010) where the emissions are driven either by specified dust emission fluxes or with simple emissions approaches based on the offline wind fields in the chemistry transport model. The model development here has been to couple the UKCA-MODE aerosol scheme to transport dust in the Unified Model driven by the existing dust emissions scheme as used by HadGEM2-ES, for example. Having the dust transported within UKCA will enable interactions with the UKCA chemistry (e.g.

heterogeneous chemistry) and may also lead to a reduced number of tracers required when dust-only no-chemistry runs are required for other purposes.

The existing UM mineral dust scheme (Woodward, 2001), calculates the emissions flux of mineral dust in several size-classes (bins) as a function of wind speed, soil moisture, vegetation and soil texture (sand, silt, clay fraction). Dust uplifted out of the saltation layer is calculated in 6 size bins with the mass in each bin carried as a separate transported tracer in the model advection scheme.

UKCA-MODE treats the aerosol size distribution in four size categories: nucleation, Aitken, accumulation and coarse modes. For each size category (except nucleation) there exists a hydrophilic and a hydrophobic mode. Of the seven modes of the UKCA scheme, the six bin-resolved dust emissions fluxes are in the size range of the accumulation and coarse modes, with an additional flux larger than about 5-10 microns, which is too large to be carried by the coarse mode and will need to be treated separately as a "super-coarse" mode. As mineral dust is hydrophobic on emission it is reasonable to directly emit the mineral dust into the accumulation and coarse hydrophobic modes.

A number of different setups have been tested to emit the bin-resolved dust into the modal UKCA aerosol scheme. At present, the configuration is that all the mass flux in bins 1 and 2 and the smaller-half of the emission flux of bin 3 are emitted into the accumulation-insoluble mode, with the larger-half of bin 3 and all of bins 4 and 5 emitted into the coarse-insoluble mode. We currently neglect the emission flux from bin 6 as, although it is a significant fraction of the total emitted dust mass, the lifetime is so short that it does not significantly contribute to mass outside of the grid box in which it is emitted. It would be possible to implement a non-advected super-coarse mode for radiation calculations if this is found to be necessary.

Evaluation of the UKCA-MODE simulated dust against that from the existing bin-resolved dust tracers has been carried out (see Figure 11) and a reasonable distribution of dust is seen from UKCA, although the dust lifetime is currently too short when compared to observations.

Modifications to the way the primary emissions (including dust) are treated in UKCA is currently underway as the current approach, which integrates the BL mixing for aerosol and then the primary emissions fluxes, is found to lead to problems, particularly for coarse particles. The UM dust scheme calculates the dust uplift based on a combined integration of the boundary layer mixing and the dust emissions, and this approach is currently being implemented into UKCA.

9 Estimation of indirect radiative effects of aerosol

A crucial link between aerosol and cloud is the ability of aerosols to act as cloud condensation nuclei (CCN) on which cloud droplets form. The first indirect effect involves increased cloud albedo from enhanced cloud droplet number concentrations (CDNC) with higher aerosol loading. The HadGEM2-ES model uses an empirical parameterization (Jones et al., 1994) to relate CDNC to the number of aerosol particles that act as cloud droplet nucleii.

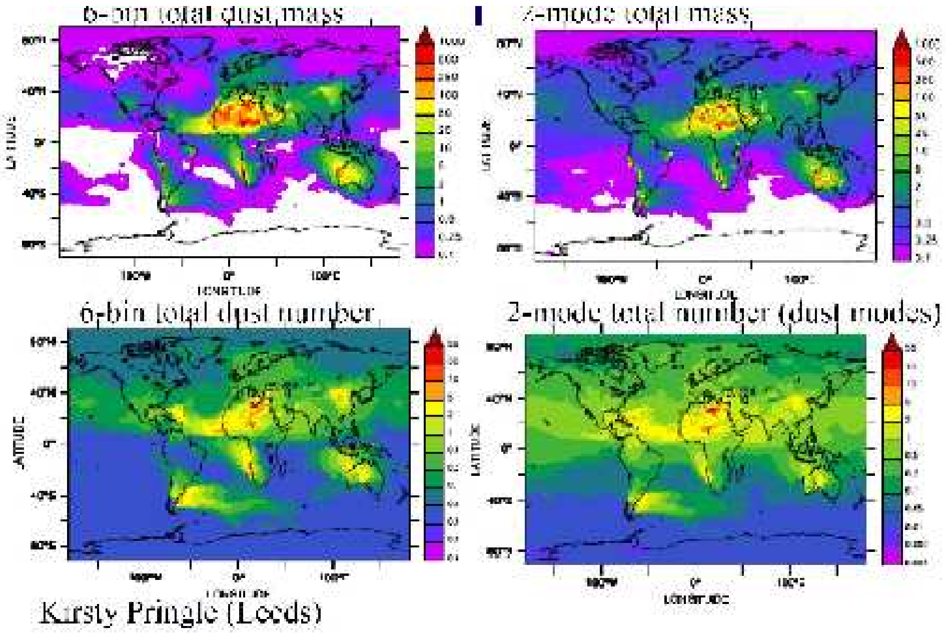

Figure 11: Example comparison of surface global maps of dust mass and number concentrations simulated in UKCA-MODE compared to those from the existing 6-bin Woodward scheme in the UM.

One of the key benefits of the UKCA-MODE scheme for UKCA is that the evolution of the aerosol size distribution is simulated dynamically, with number and mass both being caculated explicitly from the aerosol microphysics. The global CCN from UKCA-MODE will therefore be simulated more realistically than in the HadGEM2-ES model which uses the CLASSIC aerosol scheme. Figure 12 shows the number concentration of CCN that would form at a fixed supersaturation of 1 % as caculated by UKCA-MODE at each model grid point near the surface. Over plotted circles show CCN measurements mad at the same supersaturation (Spracklen, 2010).

Current development of the scheme to calculate CDNC concentrations parameterises the activation of the aerosol to form cloud droplets following the scheme of Abdul-Razzak and Ghan (2000). Variations in the local updraught velocity have a significant effect on the maximum supersaturation achieved, and hence on the number of activated particles. The sub-grid scale variability of updraught is represented using a probability density function, constrained by the turbulent kinetic energy in the boundary layer. The cloud droplet number concentration (CDNC) in warm clouds is approximated by the number of particles which activate at the cloud base. This diagnostic CDNC is then used to calculate the cloud droplet effective radius which determines the cloud albedo effect. The rate of autoconversion of cloud water to rain water is also strongly dependent on CDNC and this dependancy permits calculation of the second aerosol indirect (or lifetime) effect.

10 Deposition processes

Deposition processes to ground are important for many of the model species in the model including the aerosol component and gas-phase species such as ozone, nitric acid, and

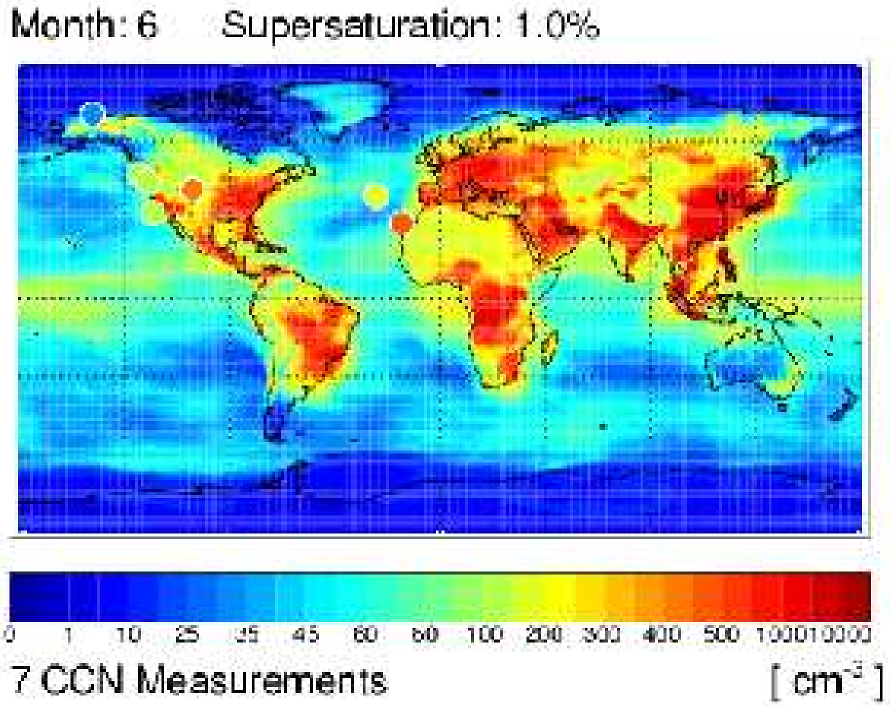

Figure 12: The number concentration of CCN that would form at a fixed supersaturation of 1 % as calculated by UKCA-MODE for June for the lowest model layer. The circles show CCN measurements made at the same supersaturation.

sulphur dioxide. The dry-deposition process is modelled using aerodynamic and surface resistances, including the stomatal conductance which is taken from the land-surface carbon cycle scheme in the Earth System models. The dry deposition scheme currently operates by extracting concentration increments throughout the boundary layer. This scheme needs modification so that the dry deposition, together with the emission fluxes are taken account of within the boundary layer scheme.

The scavenging of aerosols and soluble chemical species is also an important process for UKCA. Comparison between the global aerosol budgets in the ECHAM model (Hoose et al., 2008) and in UKCA suggest that the wet deposition process is a major source of uncertainty in the modelling of the global aerosol. In the current version of UKCA, this process is modelled through a first order loss process. The removal rate is parameterised through use of the predicted precipitation rate and cloud water content. Work to account for the sub-grid scale nature of precipitation is in progress. Work is also required to account for the re-suspension of dissolved material due to the evaporation of falling precipitation. Improvements are also needed to the convection scavenging scheme to prevent the excess transport of soluble species to the upper troposphere.

11 Code Maintainence

Recent work with the UKCA model has been on the MONSooN joint computor which is accessible to both Met Office and NERC staff. This facility has been of great benefit to the development of the project as researchers can use a common code base. Code development is maintained using the Unified Model FCM system which tracks changes. A forward workplan for merging the various development branches together has been agreed between the UKCA partners, and stage one should be completed by the end of March. This code will include the TropIsop, Stratospheric and CheST chemistries, and will include aerosol chemistry appropriate for the troposphere and stratosphere. The latest version of the UKCA-MODE will be included, together with the FastJX photolysis scheme. Another improvement to be included is a diagnostic suite for the ASAD system which allows the capture of chemical fluxes for budgeting purposes.

12 Conclusions

12.1 Chemical schemes and solver

A range of chemical schemes is now available for model development and research using UKCA, and all of these use the Newton-Raphson solver. The schemes include the CheST scheme which contains a chemistry suitable for the stratosphere and troposphere. In addition, schemes containing the chemistry required for aerosol production are also available for the tropospheric and stratospheric domains. A tropospheric heterogenous chemistry has recently been tested. The complexity of the scheme adopted for long term integrations of HadGEM3-ES will depend on the efficiency of the chemical solver and the computor resource available. Evaluation of the tropospheric chemistry model shows that the introduction of an online photolysis scheme reduces the model biases, and the addition of heterogenous chemistry also improves the model. More evaluation work will be done when the model has been consolidated. The solver is currently being optimised for the IBM computor, and it is likely that that more work will be needed in this area.

12.2 Methane simulation

As in previous models, methane may be simulated using surface emissions estimated from anthropogenic and natural sources. The accuracy of the simulation will depend on these emissions as well as on the simulated hydroxyl radical concentration. It is likely that the accuracy of the tropospheric chemistry will be improved by the use of the online photolysis scheme.

12.3 Assessment of HadGEM3 for chemical and aerosol modelling

It is important that the HadGEM3 atmosphere model provides a good platform for the UKCA model. Most work in this area has been done using the high-top version of the model with a top at 85 km. The effects of tuning the non-orographic gravity wave drag scheme are discussed above. The model intercomparison of stratospheric chemistry which included UKCA (Eyring et al., 2010) is also relevant, though it used an earlier version of the Unified Model. This activity is ongoing, with active discussions between the research institutes.

12.4 Aerosols

The adoption of UKCA-MODE should be advantageous for the HadGEM3-ES model for several reasons. Firstly, UKCA-MODE treats mixtures of different particle types (e.g. mixed soot-sulphate particles) which have been shown to be important when characterising the aerosol direct radiative forcing (e.g. Haywood and Shine, 1995). Secondly, the UKCA-MODE scheme will provide a step-change in the UK climate model capability in simulating aerosol indirect effects on climate via modified cloud albedo and lifetime. The UKCA-MODE scheme calculates the aerosol size distribution (critical to all climate influences, e.g. IPCC, 2001) as determined by the microphysical processes which control it, whereas the CLASSIC aerosol scheme used in HadGEM2-ES (in common with most first generation aerosol schemes) has to assume a globally constant size distribution for each aerosol type. The dynamically evolving size distribution in UKCA-MODE has particular benefits when simulations aerosol indirect effects (e.g. Pringle et al, 2009). Recent updates to UKCA-MODE are described here, and include new schemes to simulate stratospheric aerosol, boundary layer nucleation and mineral dust within the UKCA framework. A previous report (Johnson et al., 2010) has already described the comparison between the UKCA-MODE aerosol scheme with the CLASSIC aerosol scheme used in HadGEM2. This includes a description of the scheme for the direct radiative forcing from the aerosols included in UKCA-MODE. A new scheme for estimating the indirect effects of aerosol on clouds and precipitation is described.

12.5 Interactions

Interactions between the various components of UKCA and the UM are steadily being enhanced: for example interactions between the aerosol scheme and the chemistry now include the calculation of heterogenous chemistry rates on aerosol surfaces, and the role of aerosol in modifying photolysis rates. The added processes make the UKCA model more self-consistent, and other improvements, for example in the wet deposition of soluble aerosols and gas-phase components, will make the model more responsive to changes to clouds and precipitation which may take place as a result of climate change.

12.6 Future Work

Following the developments discussed above, the model will be consolidated so these improvements are available to all. The combined model will then be reassessed and evaluated against measurements. More work will be needed on the chemical solver in order to reduce computer costs. For UKCA-MODE, the ammonium nitrate aerosol component will be added. Recent theoretical work that may improve the local mass conservation of the UM tracer tarnsport scheme has recently been done, and the effects of this on the UKCA model will need to be investigated.

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