

UKCA Report for 2006/7

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Introduction

The UKCA project was initiated in 2004 with the objective of building and evaluating scientifically a new UK community atmospheric chemistry/aerosol model suitable for a range of applications in climate and environmental change research, integrated into the Unified Model environment. NCAS and the Met Office fund the project jointly, and the project partners are NCAS-Climate (University of Cambridge, Atmospheric Chemistry Support Unit and University of Leeds, Institute of Atmospheric Sciences), and Met Office, Hadley Centre. Most of the model development has been done using current versions of the climate model configuration at N48L38 resolution, with N48L60 resolution used for stratospheric work. Some work on regional modelling is also proceeding.

Stratospheric Chemistry

Stratospheric chemistry in UKCA has reached technical maturity and is ready for use in scientific experiments. The most recent additions to stratospheric chemistry are a state-of-the-art non-families chemical solver, which improved the model performance in the upper stratosphere and mesosphere and heterogeneous reactions on sulphate aerosol (prescribed from a climatology until superseded with UKCA interactive aerosol), which improve correspondence of modelled nitric acid with observations. It was recognized that nonlinearities in model transport led to artificial production and losses of chemical elements such as chlorine and bromine. A redefinition of tracers for transport to ensure elemental conservation has almost completely solved the elemental non-conservation problem and has led to substantial, and beneficial, increases of total odd nitrogen (figure 1).

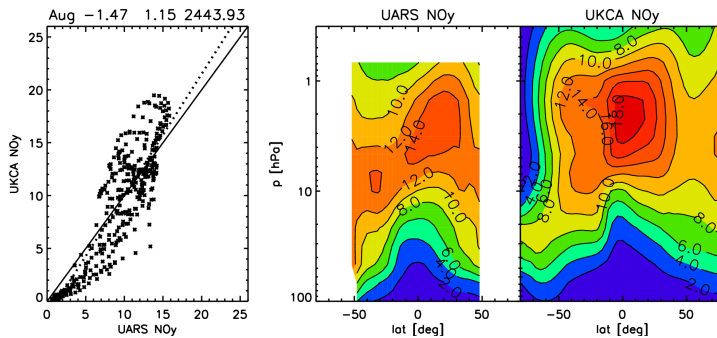


Figure 1: Zonal- and monthly mean $\text{NO} + \text{NO}_2 + \text{HNO}_3$ (ppbv) in August in a simulation with elemental conservation. (centre) UARS climatology, (right) model result (left) scatter plot of UARS versus UKCA.

A stratosphere-wide low bias of total odd nitrogen has been removed. Details of the nitrogen distribution, particularly the tongue of nitrogen-depleted air descending over the South Pole, and the underestimation of nitrogen in the extratropical lower stratosphere, point towards remaining problems with transport that need to be addressed.

A persistent problem of the stratospheric version of the New Dynamics Unified Model are global positive temperature biases in the lower stratosphere. The biases are largest over the poles and have completely suppressed heterogeneous chlorine activation on polar stratospheric clouds; consequently the model did not produce an Antarctic ozone hole. A recent model version, called here pre-HadGAM1a, has been developed which uses a new background ozone climatology, derived from a data set due to Kiehl et al. (1999). This data set has substantially more ozone at the ozone maximum, but a sharper vertical gradient and less ozone in the tropical lower stratosphere. Simulations with this ozone consequently absorb more long- and shortwave radiation in the middle stratosphere, where the impact on temperature change is small, and less radiation near the cold point, where previously there were large positive temperature biases. The warm bias is then substantially reduced and between the cold point and the ozone maximum a cold bias (figure 2) occurs.

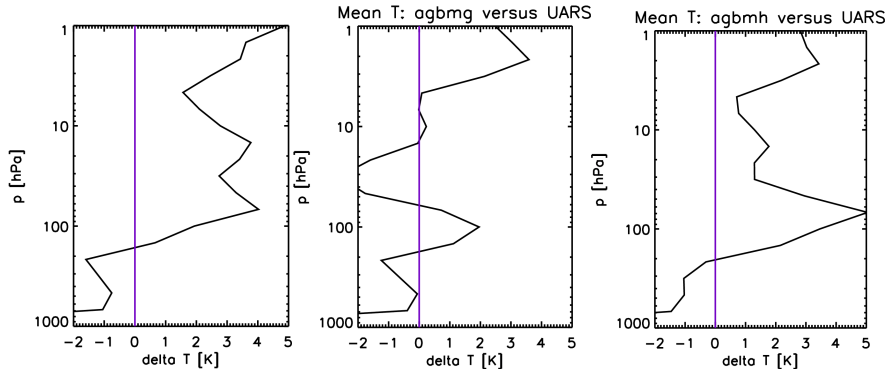


Figure 2: Global-mean temperature bias versus UARS climatology (left) with HadGAM1, using interactive chemistry; (centre) pre-HadGAM1a, using the SPARC ozone climatology; (right) pre-HadGAM1a, using interactive chemical ozone.

A three-year simulation with off-line chemistry, using this new background model, produces a more realistic ozone hole than before, indicating that when fed with appropriate temperatures, the chemistry module is indeed capable of substantial Antarctic ozone depletion (figure 3) – the model now successfully reproduces an ‘ozone hole’.

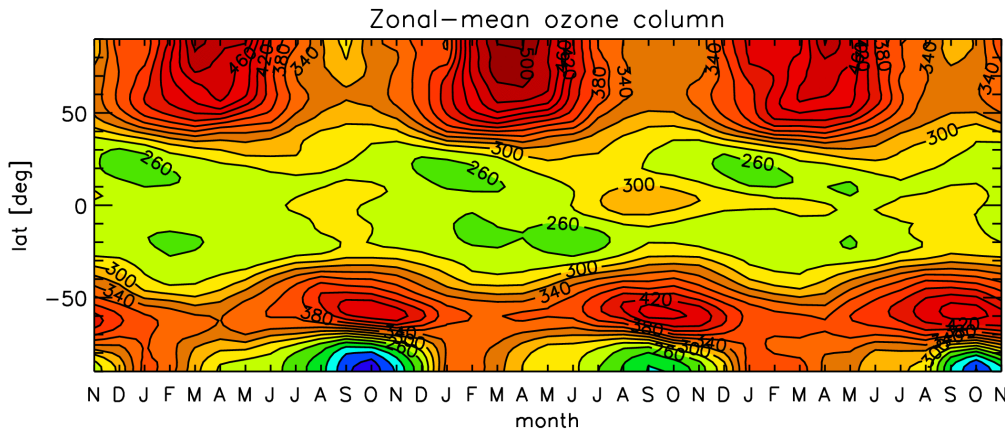


Figure 3: Zonal- and monthly-mean ozone column (Dobson units) with pre-HadGAM1a with non-interactive chemistry.

However, away from the Antarctic region tracers generally compare less favourably to climatologies than in older model versions. Further on-going model development is required.

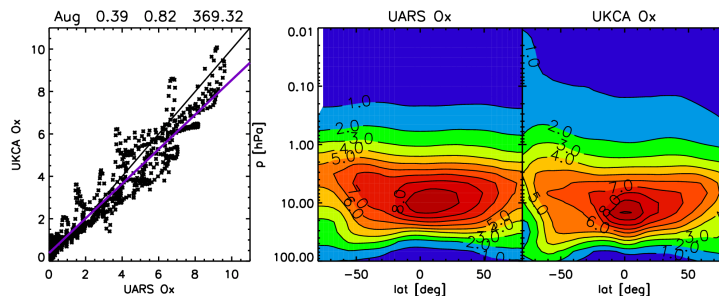


Figure 4: Zonal-mean ozone (ppbv) in August in pre-HadGAM1a.

Also, when switching to interactive chemistry the model responds by reverting largely back to the old temperature structure seen before in HadGAM1 with substantial biases in the lower stratosphere. A better understanding of the causes of the temperature bias and possibly some retuning of the model with interactive chemistry is needed to address this issue.

Tropospheric Chemistry

Chemical Budget Diagnostics

Better analysis of the tropospheric configuration of UKCA has been enabled by the addition of 3D chemical budget diagnostics for every gas-phase and photolytic reaction. Figure 5 shows zonal mean plots of the chemical production, chemical loss, and dry deposition of ozone in July. It indicates that the bulk of ozone deposition occurs in the northern hemisphere and that globally, there is net chemical destruction of ozone in the troposphere.

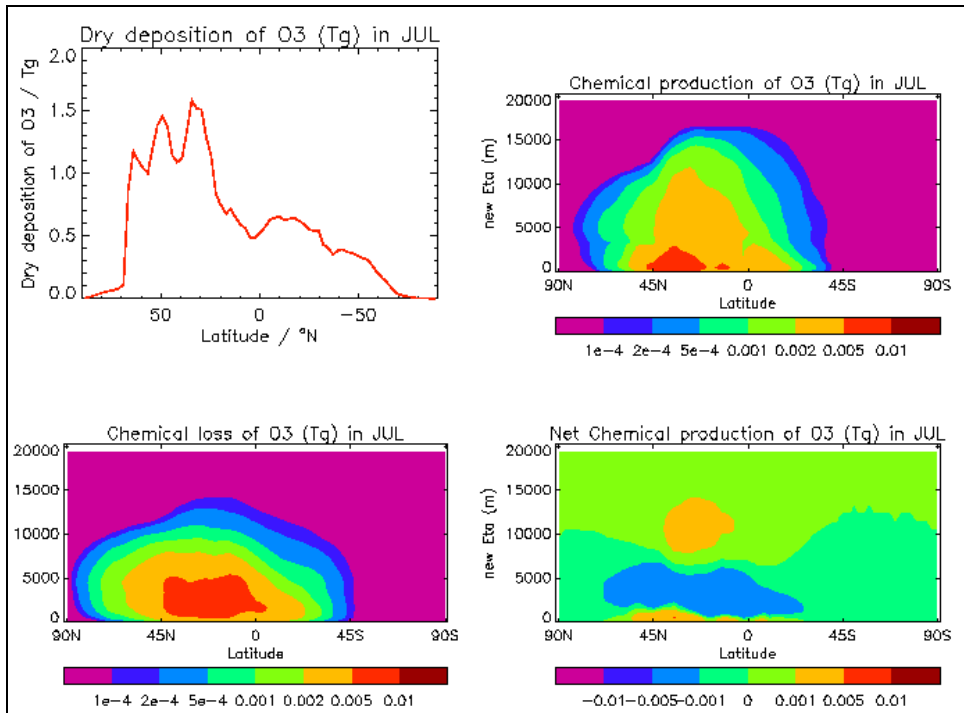


Figure 5: Zonal mean plots of the dry deposition, chemical production, chemical loss, and net chemical production of ozone for the month of July.

Coupling with Land-Surface and Vegetation Schemes

The dry deposition rates of trace gases such as ozone and nitrogen dioxide are affected by deposition to plants, and are therefore affected by changes in stomatal conductance as well as long term changes. Sanderson et al., 2007 describe a scheme linking the dry deposition of trace gases in the STOCHEM model to the MOSES surface exchange scheme of the Unified Model (Essery et al., 2001). This dry deposition scheme has now been implemented in UKCA.

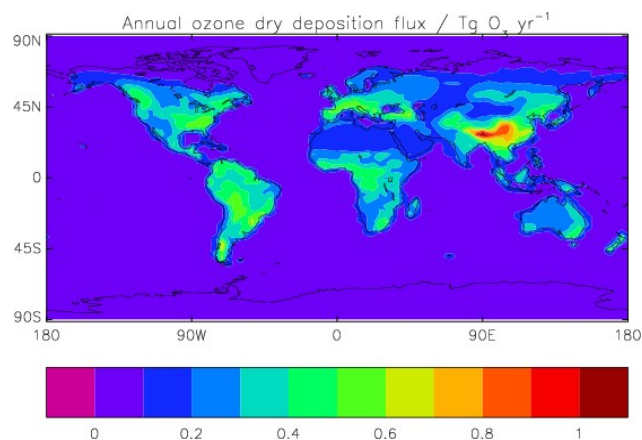


Figure 6: Annual mean ozone deposition ($Tg\ O_3 / year$) from the interactive dry deposition scheme.

The natural emissions of hydrocarbons and other substances are likely to change with both climate and vegetation changes. As a first step to incorporating these feedbacks into UKCA, the scheme of Gedney et al., 2004 to provide methane emissions from wetlands has been included.

Stratosphere-Troposphere Exchange

The chemical budget diagnostics provide specific diagnostics for the chemistry but closure of the budget can only be achieved by assuming balance and inferring a stratosphere-troposphere transport term. Explicit diagnostics were added which allow the cross-tropopause mass flux of all the chemical tracers to be calculated, thus enabling a better diagnosis of the transport in the model and closure of the chemical budgets. Figure 7 shows the stratosphere-to-troposphere mass flux of ozone from a 5-year model simulation. It indicates that the net downward transport of ozone from the stratosphere into the troposphere is too high by a factor of 3 in comparison with other tropospheric models (Stevenson et al. 2006). This could be attributed to either the cross-tropopause mass fluxes themselves being too large in the climate model or to the chemistry in the troposphere giving too strong an ozone gradient across the tropopause.

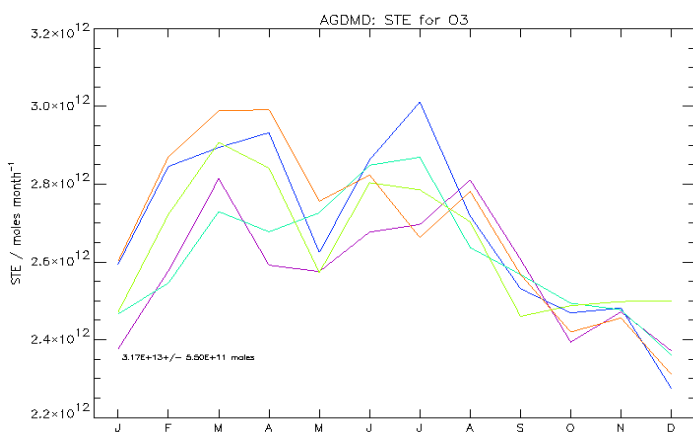


Figure 7: Seasonal cycle of stratosphere-to-troposphere mass flux of ozone from a 5-year model integration.

An assessment of the transport itself has been carried out using analysis of the transformed Eulerian annual mean residual vertical velocity at 70 hPa. Figure 8 suggests that the magnitude of the mean residual vertical velocity from HadGAM1 is within the range of other models (Butchart et al. 2006). However, the width of the tropical pipe in HadGAM1 extends too far poleward in the Southern hemisphere. Further analysis is required to assess the seasonal cycle of the transport and to examine the influence of the chemistry on the stratosphere-troposphere flux.

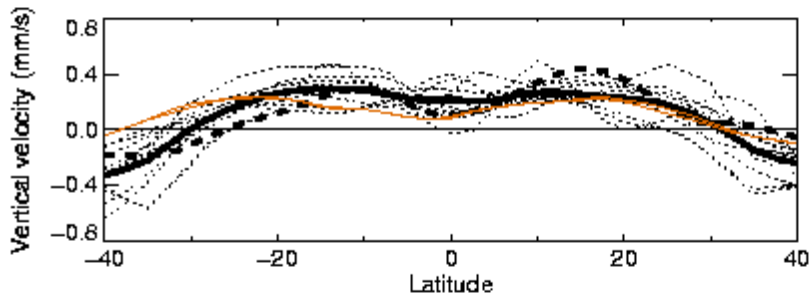


Figure 8: Annual mean residual vertical velocities at ~ 70 hPa from a multi-model ensemble from Butchart et al. 2006 (dotted). The thick line is the multi-model mean and the thick dashed line is the 10-year mean from the UK Met Office Analyses at 68 hPa. Output from 2 years of a HadGAM1 simulation are shown in orange.

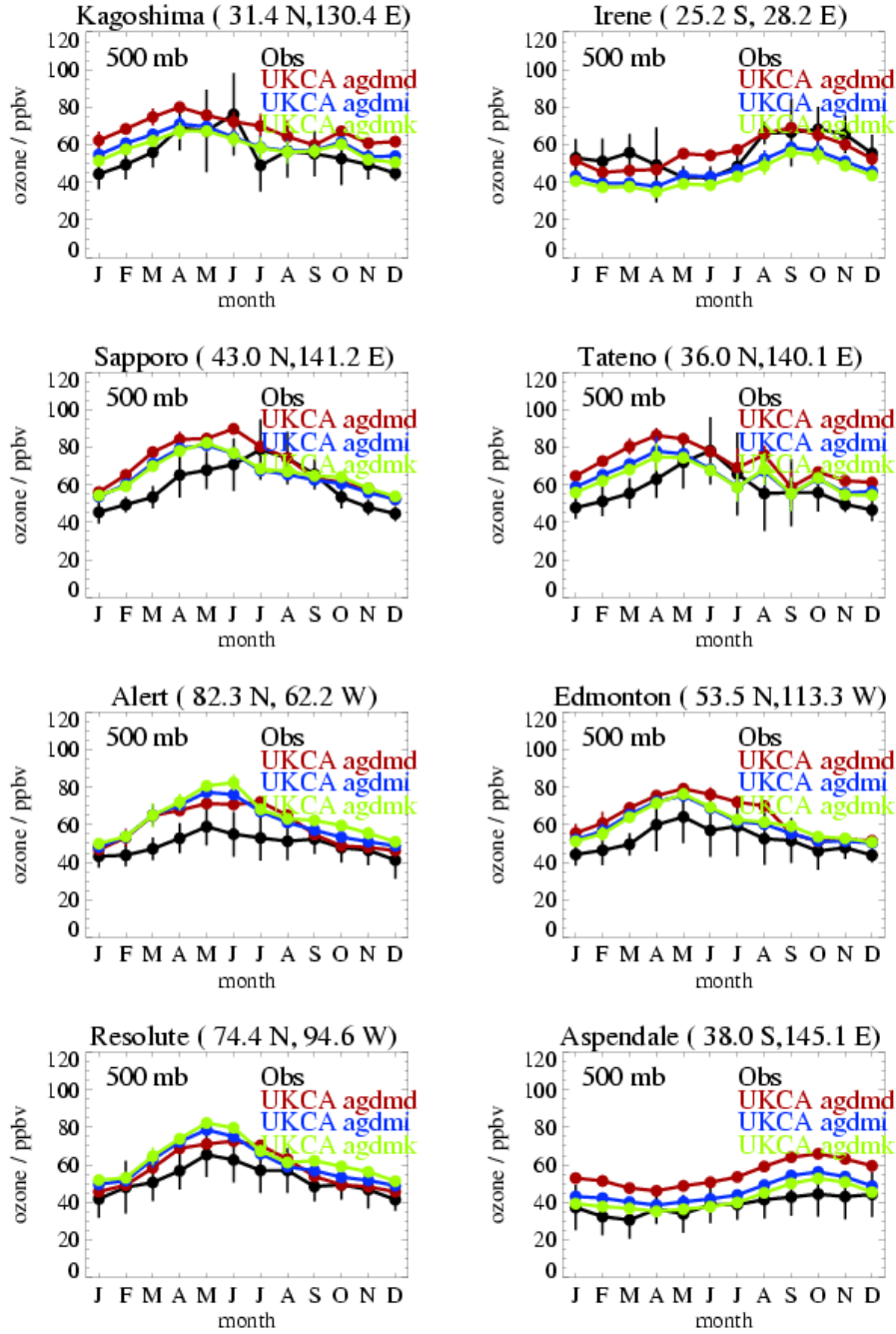


Figure 9: Seasonal variation in predicted tropospheric ozone concentrations compared to ozonesonde observations. The three model scenarios refer to the upper boundary condition for ozone in the stratosphere, with run agdmd replaced ozone at all levels in the stratosphere, run agdmi replaced ozone only above the third level above the tropopause, and run agdmk replacing ozone above the fifth level above the tropopause.

The tropospheric chemistry model requires upper boundary conditions in the stratosphere, where ozone and other long-lived trace gases are overwritten using concentrations derived from measurements or other predictions. Figure 9 compares the monthly variation in model ozone with observations for the 500 hPa level for three scenarios which vary the level above which ozone is replaced by data. This comparison is better than in previous versions of the model however, the diagnosed flux of ozone into the troposphere is still too high in this model.

Aerosol Chemistry

The tropospheric chemistry component of the model has been extended to include the oxidation of dimethyl sulphide (DMS, emitted mainly by marine phytoplankton) and sulphur dioxide (SO_2 , emitted mainly by human activities and by volcanoes). The oxidation of these species forms gas-phase H_2SO_4 which is available for nucleation and condensation processes in the aerosol model. The aqueous-phase oxidation rate of SO_2 is passed to the aerosol model directly where it is used in the growth of Aitken and accumulation mode soluble aerosols. As well as sulphur oxidation, work is in progress to model the contribution of nitrate and ammonium to the aerosol mass.

Because of the complexity of comprehensive DMS oxidation schemes (e.g. Lucas and Prinn, 2002), and reported problems with simplified chemical schemes (Lucas and Prinn, 2005), we have chosen to represent DMS oxidation in the parameterised form with product yields calculated using equations which include oxidants such as OH, NO_3 , and O_3 . Only DMS, and the products SO_2 , MSA, and H_2SO_4 are carried as species in the model, with product yields calculated from rate coefficients and oxidant concentrations. Formation of MSA and CH_3SO_3 without passing through CH_3SO_2 is included. DMS emissions to the atmosphere are calculated in the model interactively from seawater DMS concentrations using the meteorological conditions (Jones and Roberts, 2004). As an example, H_2SO_4 production from the reaction of DMS with NO_3 is shown in Fig. 10. This shows coastal intensification from both shipping and land-based NO_x emissions, and a general increase in the northern hemisphere.

Aqueous and gas-phase oxidation of SO_2 is represented. Aqueous oxidation of SO_2 by H_2O_2 has been implemented, and work is now underway to include oxidation by O_3 . In introducing aqueous chemistry, we have adopted a similar approach to that of Berglen et al. (2004). In this approach, the fraction of each species which is dissolved in cloud droplets is calculated from the temperature and cloud liquid water content via Henry's Law. The rate coefficients of aqueous reactions are then modified by multiplying by the dissolved fractions of the participating species, and by the cloud fraction. In this way, and with an appropriate conversion of units, the rates of the aqueous reactions can be used with the gas-phase concentrations of the species. In addition, where the dissolved fraction of a species is significant, the rates of any gas-phase reactions involving that species must also be modified, by multiplying by $(1 - f_d)$, where f_d is the dissolved fraction. In the chemistry component of the model, the aqueous reactions result in the removal of SO_2 but have no products. Instead, the reaction fluxes (i.e. the rates of removal of SO_2 in the

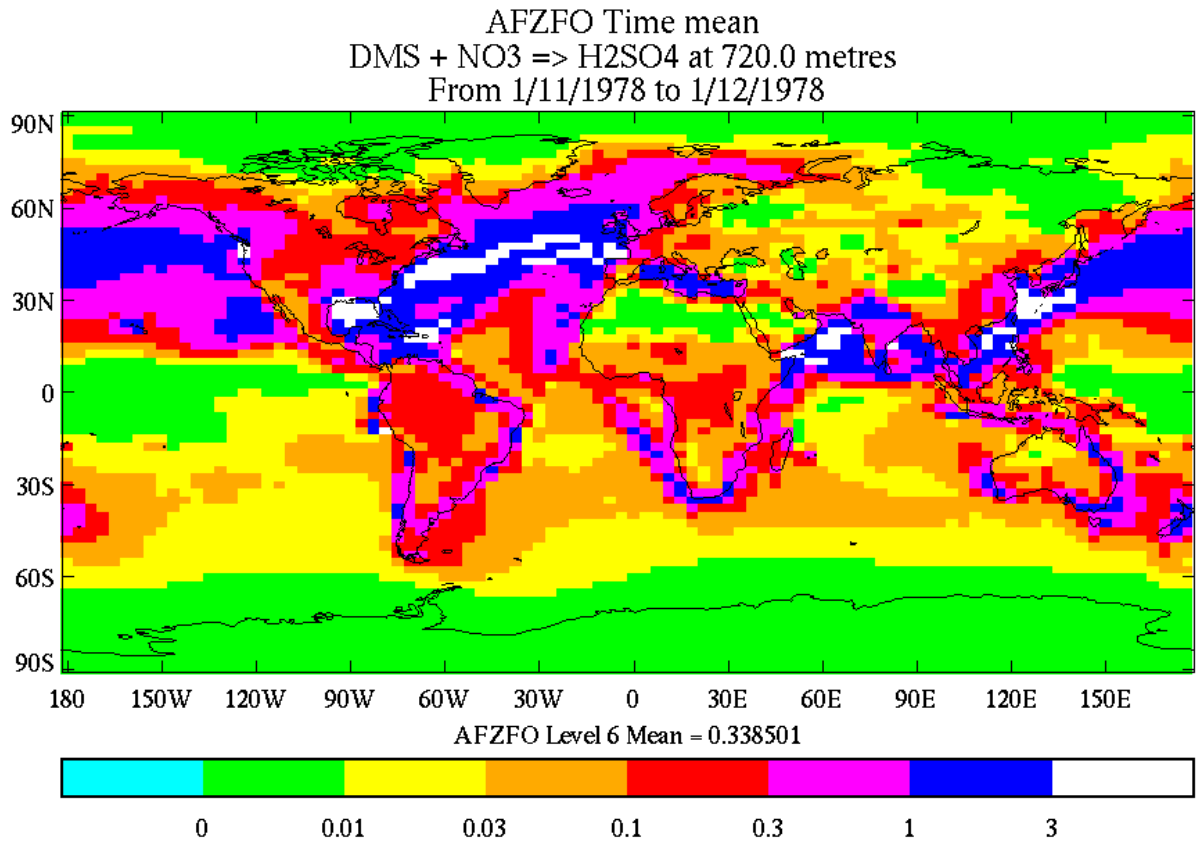


Figure 10: Rate of formation of H_2SO_4 from the reaction between DMS and NO_3 .

reactions) are passed to UKCA-mode to be used in the calculation of sulphate aerosol mass increase from cloud processing.

Aerosol Model (UKCA-MODE)

Introduction

The development of a new aerosol scheme for the UM had four major objectives:

1. An aerosol microphysics scheme that describes both particle number and mass, and thereby is able to capture changes in the particle size distribution in time and space (the existing scheme carried only mass per aerosol component)
2. The ability to simulate particles of mixed composition (important for optical properties, deposition, particle reactivity, etc.). In the existing scheme all aerosol components were assumed to be in separate particles.
3. Coupling of aerosol and gas phase chemistry (required to predict changes in particle composition and to use online oxidants).
4. To include major components of the aerosol hitherto neglected: secondary organic material and the capability to simulate the complete inorganic composition ($\text{H}^+/\text{Na}^+/\text{NH}_4^+/\text{SO}_4^{2-}/\text{NO}_3^-/\text{Cl}^-$)

Summary

- The new aerosol scheme being implemented for UKCA (named “UKCA-mode”) has been developed and tested. It has a complete treatment of the microphysics and represents the particles as a series of dynamic log-normal modes with particle number and amounts of each chemical component as prognostics.
- The scheme currently includes prognostic sea spray and anthropogenic and natural black carbon, organic carbon, secondary organic carbon and sulphate.
- A version of UKCA-mode has now been successfully ported to the UM and is currently being evaluated (to speed up code development and testing, UKCA-mode was originally implemented within the TOMCAT offline CTM)
- Two versions of the code have been developed: one that can separate soluble and insoluble particles (requiring 25 aerosol advected tracers) and one that lumps all components together in mixed soluble particles (18 tracers). Multicomponent inorganic aerosol will add additional tracers. This number of tracers is comparable to the intermediate gas phase chemical scheme.

- Ongoing evaluation suggests that UKCA-mode is performing well and is sufficiently fast for 100 year simulations, as planned.
- UKCA-mode is part of several observational or modeling collaborations. The development of inorganic composition code (item 4) and a new secondary organic scheme are part of these collaborations. represents a significant improvement over existing capability in the Unified Model. UKCA-mode implements the GLOMAP aerosol microphysics (Spracklen et al, 2005a, 2005b, 2007) within a modal aerosol dynamics framework similar to the HAM aerosol model (Stier et al, 2005) which was developed for the ECHAM. Such modal schemes are more GCM consistent with the CPU constraints of a General Circulation Model than the GLOMAP sectional scheme.

Improvements in aerosol modeling capability that UKCA provides are i) a representation of particle growth (due to condensation, coagulation, cloud-processing) is included by using a 2-moment (number and mass) modal scheme ii) a new particle formation scheme populates the various aerosol size modes more physically, iii) on-line coupling with gas phase chemistry to predict the aerosol response to changes in atmospheric oxidizing capacity, iv) a representation of internally mixed aerosol which feeds into the UM radiation scheme, v) a simple secondary organic aerosol scheme.

Collaborations

A major change since the last Board Meeting is the rapidly increasing number of projects contributing to aspects of UKCA-mode development, evaluation or exploitation. UKCA involvement in national and international research projects will add substantially to the evaluation effort, but is adding to the workload of the UKCA team in Leeds and requires careful coordination.

Outside Leeds:

- Figure 1 shows how the future versions of UKCA-mode are likely to improve following ongoing projects funded by NERC and the Met Office involving UKCA and GLOMAP. These include improvements in aerosol The thermodynamics of inorganic mixed aerosol (including nitrate and ammonium aerosol) was supported by Manchester's DIAC work. They have produced a look-up table to speed up the solution of the inorganic composition equations. This will be implemented as part of the (QUEST-ESM project
- New schemes for secondary organic aerosol are progressing as part of the QUEST-QUAAC project. Leeds have made an appoint-

ment in this project and will now begin testing several SOA scheme in UKCA

- The European Integrated Project (EUCAARI) plans to use UKCA as part of the Earth System modeling work package. Leeds are leading a WP on parametrisation development, which aims to improve modal schemes as used in UKCA-mode.
- In APPRAISE, Edinburgh (Palmer) proposes to use GLOMAP and UKCA to study organic aerosol.

Projects in Leeds specifically planning to use UKCA:

- The role of heterogeneous chemistry is being investigated by (Mat Evans through a CASE studentship with the Met Office)
- The representation of marine aerosol is being investigated as part of a (SOLAS project, a SOLAS tied studentship (halogen/sulphur cycle) and a Met Office & tied, CASE studentship (dust and DMS in the Earth System).
- The effect of particle formation on global CCN is part of the AEROFORM NERC project
- The UKCA aerosol optical depth is being studied through a NERC DIAC studentship and the APPRAISE-ADIENT consortium.
- Stratospheric aerosol will be studied through a NERC collaboration of Pyle and Carslaw.

Taken as a whole these advances will further enhance the realism of Hadley Centre simulations of aerosol impacts on climate and the earth system.

Status and Progress

Model development: UKCA-mode has been developed and tested within the offline chemistry transport model TOMCAT (Stockwell & Chipperfield, 1999). There are several advantages to this approach: (i) TOMCAT is faster and easier to use and adapt than the UM; (ii) the CTM uses analysed winds, making comparison with some observations easier; (iii) UKCA-mode can run alongside the more complex model GLOMAP (which uses >20 size sections and >100 tracers), which can serve as a benchmark model. This approach has proved successful. We now have two advanced aerosol models within the same CTM and there is increasing interest from the community to use such an offline aerosol CTM. All work with the CTM will result in an improved UM aerosol scheme.

The microphysics code handling the emission, growth, coagulation, cloud processing and deposition of particles is essentially complete. It currently includes sea spray and primary anthropogenic and natural carbonaceous particles as well as secondary sulphate and organic

material. Example fields of particle number and cloud condensation are shown in Figure 11. A new scheme for multicomponent inorganic aerosol and a more advanced scheme for secondary organic aerosol can now be readily incorporated. Dust transport and deposition has been tested in GLOMAP and will soon be transferred into the modal framework of UKCA-mode.

Porting of a sulfate-and-seasalt-only version of the UKCA-mode code to the UM system began in July 2006. An interface from the main UKCA module to the UKCA-mode routine has been coded up and initial testing is ongoing. Problems gaining remote access from Leeds to the Met Office mean that this aspect of the work progressed more slowly than expected in the 2006 UKCA report to the board. Leeds will shortly have a broadband link to the Met Office.

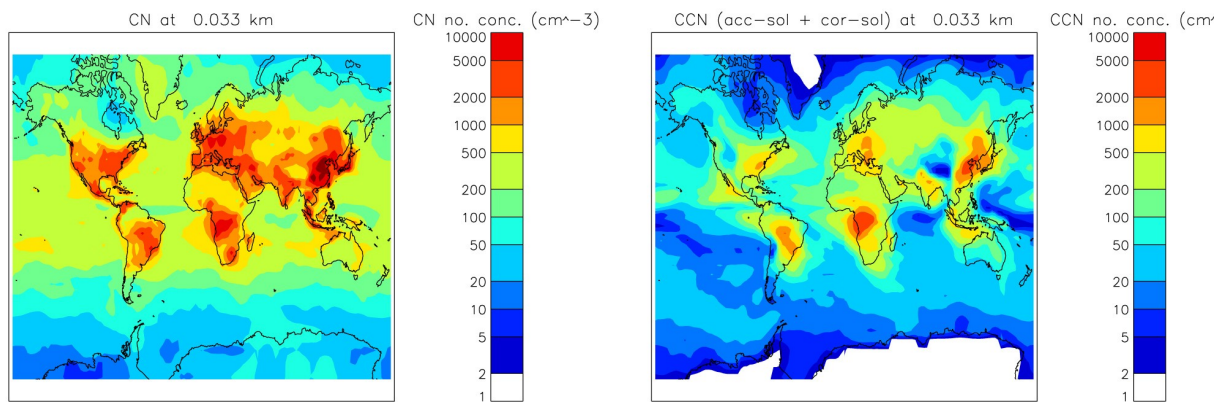


Figure 11: UKCA-mode condensation nuclei (total particle number) and cloud condensation nuclei. Particle number concentration is a new feature of UKCA-mode compared to the previous UM aerosol scheme. The model will enable cloud condensation nuclei concentrations to be calculated on a physical basis in terms of the particle size distribution and composition, rather than relying on empirical fits to observational data.

Model evaluation: New size-resolving models like UKCA-mode require new evaluation datasets that go beyond those used in previous international assessments like AEROCOM. A comprehensive observational dataset is being accumulated at Leeds for the ongoing evaluation of GLOMAP and UKCA (including size-resolved Aerosol Mass Spectrometer data recently compiled by J. Jimenez at Boulder; CCN data from over 20 years of publications; particle number data from >20 long-term observation sites; size distributions from European sites, etc.). We will also have access to extensive data through the EUCAARI EU Integrated Project to be archived at NILU.

Evaluation of a version of UKCA-mode which includes sulfate, seasalt, black carbon and organic carbon has been carried out for a year-2000 simulation within TOMCAT. The model has so far been evaluated against observations of SO_2 and $\text{PM}_{2.5}$ SO_4 over Europe (EMEP network) and of $\text{PM}_{2.5}$ SO_4 , BC and OC over the US (IMPROVE network). The model does a good job at representing SO_2 and sulfate over the US and Europe on an annual mean basis but tends to underestimate sulfate during NH winter, perhaps as a result of the omission of ammonia and heterogeneous chemistry effects. BC over the US is underestimated. However, some studies have suggested that the Bond et al (2004) primary BC emissions inventory underestimates true emissions. The annual mean OC comparison is encouraging, but winter OC is underestimated. The model SOA dominates the OC burden over the US during summer. Figure 11 shows examples of model evaluations against measurements.

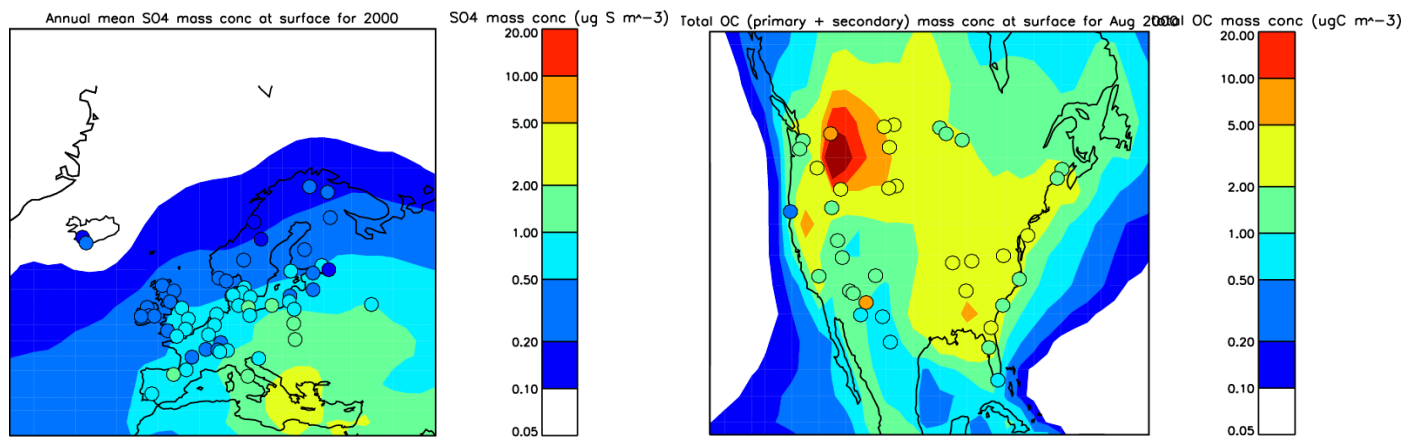


Figure 12: Examples of model evaluation. Left – annual mean sulphate at the surface for year 2000. Right – carbonaceous aerosol during the wildfire season, August 2000.

Global mean burdens and budgets have been calculated for the UKCA-mode scheme in TOMCAT. The intra-annual variability in the global burdens of SO_4 , BC, primary OC and secondary OC is within the range of values reported for other global aerosol models.

A paper using UKCA-mode to investigate changes in global and regional sulfate aerosol since the 1980s has been accepted in GRL (Manktelow et al., 2007 in press).

Tracer Transport in the Unified Model

The Unified Model incorporates a semi-Lagrangian tracer transport scheme which is used for the advection of chemical species. Early experiments with the model used simulations of Radon-222 (half life of 3.8 days) to demonstrate that global conservation was maintained. In addition to global conservation, stratospheric experiments require the conservation of chemical elements because they set the longest chemical timescales in the model. Figure 13 depicts total chlorine, initialized to 3.5 ppbv globally and maintained at this mixing ratio at the surface, during the course of a one-year model simulation.

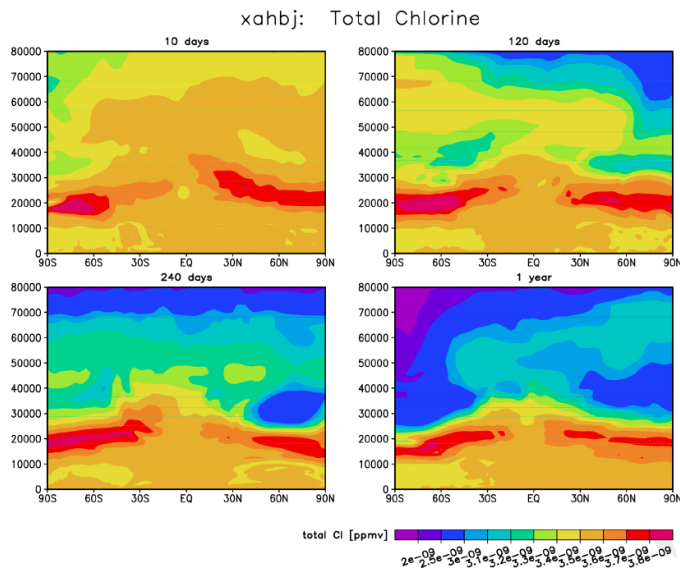


Figure 13. Zonal-mean total chlorine volume mixing ratio after 10, 120, 240, and 360 days. Figure courtesy of Margaret Hurwitz.

In this simulation, in which every tracer represents one individually advected species, in parts of the atmosphere nearly half the total chlorine is lost; in the UTLS regions, there are some gains. Since there are neither sources nor sinks for total chlorine in this model, both losses and gains are numerical artefacts. This behaviour is due to non-linearities in the transport scheme, caused by enforcement of monotonicity and global mass conservation. We established that when the transport is linearized, chemical elements that are uniformly distributed in the model are conserved. However, without global mass conservation large drifts occur that make longer runs impossible. It was therefore decided that the default settings for transport should remain to be used for UKCA and elemental conservation would be achieved by redefining tracers such that chemical elements (meaning here Br, Cl, N, and optionally, with chemically interactive water vapour, H) appear as tracers. With this redefinition multiannual

simulations are possible that do not suffer from intolerable artificial drifts in chemical elements. However, this can impact the partitioning between species within the specific chemical elements and further development is required.

Current work at the Met Office includes incorporation of the SLICE (Semi-Lagrangian Inherently Conserving and Efficient) scheme into the Unified Model. This scheme is described by Zerroukat, Wood and Staniforth (2004). Currently this code is being tested with idealised flow problems. It is anticipated that the problem with elemental conservation described above will be resolved with the implementation of SLICE.

Nudged Model Development

The development of a nudged model is important as such a model will improve the ability of the model to utilise campaign data and will also show the role of biases in the underlying model.

We have developed a version of the UKCA model in which fields of u , v and T are nudged towards the ERA-40 analyses. A test run has completed one year with no signs of instability. Preliminary results demonstrate that the model both removes biases (Figure 14 shows temperature biases in the free-running and nudged models, versus ERA-40) and is able to track variability. This improvement has been seen in the variables that are directly nudged (u , v , T) as well as in non-nudged variables (P_0 , humidity, etc.).

At present a full assessment of model performance is being carried out.

The nudged model promises to be a powerful tool for chemical process studies,

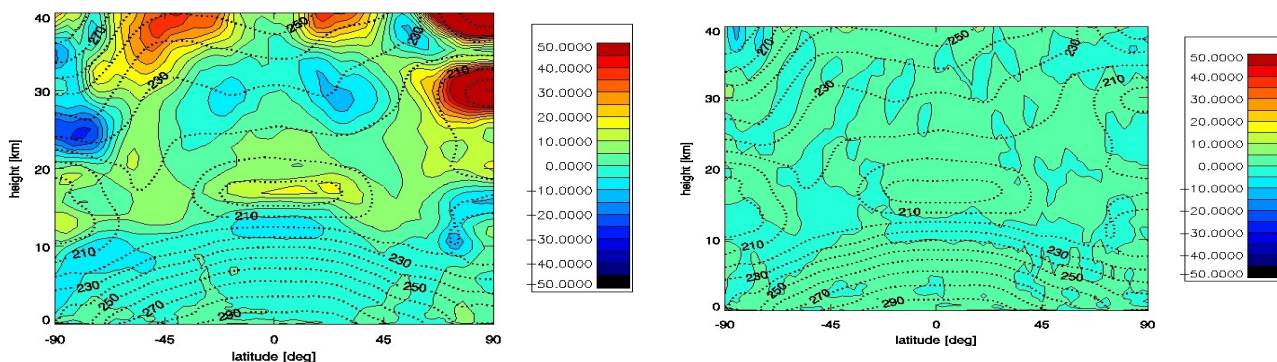


Figure 14: Difference in potential temperature (K) between the free running UKCA model and ERA-40 for October 30 1999 (left) and between the nudged model and ERA-40 (right). Dashed lines are isotherms from the analyses.

Creation and Maintenance of the UKCA Model

The UKCA model is designed so that it can be integrated into the Unified Model (UM) system, and controlled by panels in the Unified Model User Interface (UMUI). Most of the work with the UMUI panels and control files has been completed but the full system has not yet been tested. One problem has been the difficulty of working on one code when the model is being developed in three different centres, as access to the Met Office computers from outside has been difficult until quite recently. Lack of conformity between the UM versions being used, the underlying climate model version, and computer systems can all create problems in model development. For several reasons UKCA has not yet become a maintained component of the UM, though this is vital if the UKCA project is to succeed. However the system tasks involved with the creation and maintenance of a complex community model are considerable.

The development of the UKCA model has been seriously affected by problems caused by erroneous tracer transport, with subsequent loss of local mass conservation. Another problem has been the stratospheric temperature biases in the L60 model which has been under development at the same time as UKCA. These problems are likely to be resolved more quickly if tuning of the base climate models was performed taking into account the requirements important for UKCA. This requires closer cooperation between the UKCA team and development groups within the Met Office than is currently the case.

With the probability of use and development of the UKCA model involving more institutions in the future, a new phase in the project has been reached. The project partners need to address the future management of the model in this new environment. In particular attention must be focussed on the following aspects:

- How to maintain a strong central code while use of the code

diversifies

- The need to provide sufficient effort for system maintenance tasks
- The need to strengthen links between UM developers (e.g. in the dynamics area) and UKCA scientists
- To devise an inclusive management structure

Other Relevant Projects

The UKCA model is being used as a component of two Earth-System models: HadGEM2-ES and QUEST. HadGEM2-ES is the Hadley Centre Earth-System model with an interactive carbon cycle model. This model will shortly be frozen, and will contain the UKCA tropospheric chemistry model in order to provide oxidants on-line for use in the UM aerosol scheme, and ozone and methane concentrations. UKCA is also being used in a regional model at the Met Office to evaluate it against GEMS project verification data. Furthermore, the UKCA model will be used in the ongoing European SCOUT-O3 project, which focuses on stratospheric and upper-tropospheric ozone, and in SPARC's CCMVal; in SCOUT-O3 UKCA will be used to support both global and regional studies.

Conclusions

The UKCA model has been developed as a component of the Unified Model, and is being tested in a number of configurations: 1) A stratospheric chemistry model using N48L60 resolution; 2) A tropospheric chemistry model at N48L38 and N96L38; and 3) A new aerosol model at N48L38. The development of these models has exposed problems with the tracer transport scheme in the UM, and underlines the importance of collaboration between modellers responsible for different components of the Earth-System model.

Challenges for the future include the production and development of UKCA within the UM system, and the continued scientific development of the model. There is a high level of interest in the use of UKCA, and we look forward to an increased use of the model to address a range of questions concerned with chemistry and aerosol

modelling.

Acknowledgements

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References

Berglen T.F., Berntsen T.K., Isaksen I.S., and Sundet K., A Global Model of the Coupled Sulphur/Oxidant Chemistry in the Troposphere: The Sulphur Cycle, *J. Geophys. Res.*, 109, D19310, doi:10.1029/2003JD003948, 2004.

Bond, T. C.; D. G. Streets, K. F. Yarber, S. M. Nelson, J.-H. Woo and Z. Klimont, A technology-based global inventory of black and organic carbon emissions from combustion, *J. Geophys. Res.*, 109, D14203, doi: 10.1029/2003JD003697 , 2004.

Butchart, N., et al., Simulations of anthropogenic change in the strength of the Brewer-Dobson circulation, *Clim. Dyn.*, 27:727-741, DOI 10.1007/s00382-006-0162-4, 2006.

Dentener, F.; S. Kinne, T. Bond, O. Boucher, J. Cofala, S. Generoso, P. Ginoux, S. Gong, J. J. Hoelzemann, A. Ito, L. Marelli, J. E. Penner, J.-P. Putaud, C. Textor, M. Schulz, G. R. van der Werf, and J. Wilson, Emissions of primary aerosol and precursor gases in the years 2000 & 1750 prescribed data-sets for AeroCom, *Atmos. Chem. Phys.*, 6, 4321-4344 ,2006.

Essery, R., Best, M. and Cox, P. 2001. *MOSES 2.2 Technical Documentation*, Hadley Centre Technical Note No.30, 2001.

Gedney, N., Cox, P.M., and Huntingford, C., Climate feedback from wetland methane emissions, *Geophys. Res. Lett.*, 31, L20503, doi:10.1029/2004GL020919, 2004.

Jones, A., and Roberts, D.L., *An Interactive DMS Emissions Scheme for the Unified Model*, Hadley Centre Technical Note No. 47, 2004.

Kiehl, J. T., Schneider, T. L., Portmann, R. W., and Solomon, S., Climate forcing due to tropospheric and stratospheric ozone, *J. Geophys. Res.*, 104, D24, 31,239-31254, 1999.

Lucas, D.D., and Prinn, R.G., Mechanistic studies of dimethylsulfide oxidation products using an observationally constrained model, *J. Geophys. Res.*, 107, D144201, doi:10.1029/2001JD000843, 2002.

Lucas, D.D., and Prinn, R.G., Sensitivities of gas-phase dimethylsulfide oxidation products to the assumed mechanisms in a chemical model, *J. Geophys. Res.*, 110, D21312, doi:10.1029/2004JD005386, 2005.

Manktelow, P., G.W. Mann, K.S. Carslaw, and M.P. Chipperfield , Regional and global trends in sulphate aerosol since the 1980s, *Geophys. Res. Lett.*, in press, 2007.

Sanderson, M.G., Collins, W.J., Hemming, D.L., and Betts, R.A., Stomatal conductance changes due to increasing carbon dioxide levels: Projected impact on surface ozone levels, *Tellus B*, 59, 404-41, 2007.

Spracklen, D. V.; K. S. Carslaw, M. Kulmala, V.-M. Kerminen, G. W. Mann, and S.-L. Sihto, "The contribution of boundary layer nucleation events to total particle concentrations on regional and global scales", *Atmos. Chem. Phys.*, vol. 6, no. 12, pp. 5631-5648 (2006)

Spracklen, D. V.; K. J. Pringle, K. S. Carslaw, G. W. Mann, P. Manktelow, and J. Heintzenberg, Evaluation of a global aerosol microphysics model against size-resolved particle statistics in the marine atmosphere, *Atmos. Chem. Phys.*, 7, 2073-2090, 2007.

Stevenson, D.S. et al., Multimodel ensemble simulations of present-day and near-future tropospheric ozone, *J. Geophys. Res.*, 111, D08301, doi:10.1029/2005JD00638, 2006.

Stier, P., J. Feichter, S. Kinne, S. Kloster, E. Vignati, J. Wilson, L. Ganzevald, I. Tegen, M. Werner, Y. Balkanski, M. Schulz, O. Boucher, A. Minikin, and A. Petzold, The aerosol-climate model ECHAM5-HAM, *Atmos. Chem. Phys.*, 5, 1125-1156, 2005.

Stockwell, D. and M. P. Chipperfield, A tropospheric chemical transport model: Development and validation of the model transport scheme, *Q. J. R. Meteorol. Soc.*, 125, 1747-1783, 1999.

Zerroukat, M., Wood, N., and Staniforth, A., A monotonic and positive-definite filter for a semi-Lagrangian Inherently Conserving and Efficient (SLICE) scheme, *Q. J. R. Meteorol. Soc.*, 131, 2923-2936, 2005.

APPENDIX: The UKCA-mode aerosol scheme.

The set-up of the particle modes in UKCA-mode is shown in Table 1. The 5 aerosol components (black carbon (BC), organic carbon (OC), SO₄, dust (DU) and sea salt (SS)) are carried in 7 externally-mixed log-normal modes which represent differences in particle size and water-solubility. The 3 insoluble modes (Aitken, accumulation, coarse) represent i) freshly emitted BC/OC particles from anthropogenic and biomass burning sources (Ait-ins), ii) freshly emitted small mineral dust particles (acc-ins), and iii) freshly emitted large mineral dust particles (cor-ins). All particles in the insoluble modes are assumed to be hydrophobic. The 4 soluble modes (nucleation, Aitken, accumulation, coarse) represent i) particles produced from binary H₂SO₄-H₂O nucleation (nuc-sol), ii) primary sulfate particles, aged BC/OC particles and particles which have grown/coagulated from the nuc-sol mode (Ait-sol), iii) smaller sea-salt particles, smaller aged dust particles and particles which have grown from the Ait-sol mode or coagulated from other modes (acc-sol), iv) larger sea-salt particles and particles which have grown from the acc-sol or coagulated from other modes (cor-sol).

Table 1. Set-up of UKCA-mode. 25 tracers are required to simulate the number of particles within each mode and the mass of each chemical component within the mode. This reduces to 18 if the soluble and insoluble modes are combined.

Mode name	Radius range (nm)	Composition	Production	Comments
nucleation soluble	< 5	SO ₄	nucleation	
Aitken insoluble	5 – 50	BC, OC	primary emissions	Separation necessary if ageing > lifetime in grid box
Aitken soluble	5 – 50	BC, OC, SO ₄	condensation on Aitken insoluble, coagulation	

Accum. insoluble	50 – 500	DU	primary emission	Separation to handle dust ageing
Accum. soluble	50 – 500	BC, OC, SS, DU, SO ₄	growth of Aitken soluble, sea spray emission, coagulation	Primary accum. mode BC/OC ageing?
Coarse insoluble	>500	DU	primary emission	Separation to handle dust ageing
Coarse soluble	>500	BC, OC, SS, DU, SO ₄	growth of accum soluble, coagulation	

Mass is added to these modes (whilst conserving number) due to i) in-cloud oxidation of SO₂ by H₂O₂ and O₃, ii) condensation of H₂SO₄ and SEC_ORG vapour, iii) inter-modal coagulation, iv) heterogenous production of sulfate and nitrate aerosol.

A “reduced” version of UKCA-mode is also in development which carries the 5 aerosol components in just the 4 soluble modes, omitting the separation between fresh and aged dust and carbonaceous aerosol. The reduced version will require the advection of 18 aerosol tracers compared with compared with the full version’s 25. Note that the equations for the microphysics are solved process-split with analytical solutions. Consequently, tracer advection will likely be the limiting factor in computational speed.