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Deliverable Title: UKCA merged code implemented and tested ready to form part of version 8.2 of the Unified Model.

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Summary for non-technical audience

1) The UKCA community model now contains major upgrades to the chemical schemes, photolysis calculation, aerosol scheme, and the cloud droplet number concentration prediction scheme. A large number of smaller changes have also been made.

2) The new UKCA code is required in order to start work on the HadGEM3-ES model, and it is anticipated that this will use UKCA to provide a prediction of ozone in the entire model domain to 85 km, and use the MODE aerosol scheme to provide the aerosol forcings in the atmosphere model.

3) The various research branches of UKCA have recently been combined in a single branch and this code will form part of the next version of the Unified Model.

4) A recent assessment of the aerosol optical depth predicted by the UKCA model has recently been done against AERONET sun photometer measurements, and the UKCA model showed a better performance than the CLASSIC aerosol model predictions.

5) A comparison was made of the direct and first indirect anthropogenic aerosol forcing predicted by UKCA and CLASSIC between 1860 and 2100. For the all-sky forcing, where both direct and first indirect forcings are active, both aerosol modules produce the same forcing of -1.7 Wm^{-2}. This agreement on the global average masks important regional differences between the two models. In particular, over the maritime stratocumulus regions off the shore of Namibia, Peru, and California, UKCA-MODE aerosols exert weaker (less negative) forcings. This is a consequence of the assumed link between aerosol mass and number in the CLASSIC model compared to the explicit modelling of aerosol number and mass in UKCA-MODE.

Policy Relevance

It is important to improve the model predictions of radiation forcing by trace gases and aerosols in the atmosphere. Here we describe improvements to the UKCA chemistry and aerosols model which will form part of the next Earth Sytem Model.

Press/Public Interest

None.

Contents

1 Introduction

Uncertainties in radiative forcing from aerosols are documented in the IPCC Fourth Assessment Report (Solomon et al., 2007). The contribution to the overall uncertainty from aerosol components remains high, and this is the key reason driving the development of better aerosol models. Anderson et al. (2005) discuss the uncertainties in direct climate forcing from aerosols, and consider that these uncertainties could be substantially reduced by utilising satellite and other recent measurements to constrain and validate model predictions. Clearly, improvements to modelling aerosol processes are important, and the UKCA-MODE aerosol scheme offers clear advantages over older schemes, notably by treating both aerosol number and mass concentrations as prognostic variables. In addition to the aerosol forcing, radiative forcing from trace gases such as methane, ozone, and chloroflourocarbons is still important, and the prediction of future ozone concentrations from a model encompassing both the stratosphere and troposphere is desirable.

Here we discuss recent developments in the UKCA model which are currently being prepared for submission to the UM trunk and will form part of the next release (version 8.2) of the Unified Model (MetUM). These improvements are required for the development and testing of the Earth System model (HadGEM3-ES). It is anticipated that the MODE aerosol scheme will be the default scheme in HadGEM3-ES, replacing CLASSIC. With the adoption of a high-top (85 km) atmosphere domain, the prediction of ozone over the entire domain requires both stratospheric and troposopheric chemistry to be active. Other new components to UKCA include: a consistent set of chemical mechanisms for tropospheric and stratospheric chemistry; the FastJ-X photolysis scheme; tropospheric heterogenous chemistry; a scheme to predict stratospheric aerosol; and a boundary layer nucleation scheme for MODE. Note that an earlier report (Abraham et al., 2011) described in detail some of the developments to UKCA which are now being implemented at version vn8.2.

2 The HadGEM3 Climate Model

The United Kingdom Chemistry and Aerosol (UKCA) model is being coupled to the HadGEM3 Hadley Centre climate model, which is based on the Met Office Unified Model (MetUM). HadGEM3 is being developed with a variety of resolutions, here the horizontal latitude-longitude grid in use is a staggered Arakawa C-grid (Arakawa and Lamb 1977) with a resolution of N96 (1.25° latitude x 1.875° longitude). A staggered Charney-Phillips grid is used in the vertical with 85 levels extending up to 85 km. The dynamical timestep corresponding to this resolution is 20 minutes. The configuration used for the integrations described here was the atmosphere-only version of HadGEM3, with sea-surface temperatures and sea-ice prescribed as seasonally varying fields.
Coupled within the climate model, UKCA uses components of the MetUM for the large-scale advection, convective transport, and boundary layer mixing of its chemical tracers. The large-scale transport is based on the new dynamical core implemented in the MetUM by Davies et al. (2005). Advection is semi-Lagrangian with conservative and monotone treatment of tracers. Convective transport is treated according to the mass-flux scheme of Gregory and Rowntree (1990) and is applicable to moist convection of all types (shallow, deep, and mid-level) in addition to dry convection. For boundary layer mixing, UKCA uses a new boundary layer turbulent mixing scheme (Lock et al. 2000) which includes a representation of non-local mixing in unstable layers and an explicit entrainment parameterization.

3 Overview of the UKCA model

The UKCA model is a framework program capable of running a variety of chemistry and aerosol models. It is being used for both global and limited-area simulations. This report describes the status of the UKCA model. During 2010, the UKCA model has been consolidated into a single branch at version 7.3 of the MetUM, and this model has been merged in with the current (version 8.1) code in the MetUM trunk. This will be the basis for updating the trunk to form the new version 8.2. The UKCA code now consists of around 200 subroutines with over 91000 lines. The following list of the main model components includes a short description of the new capabilities which will be available at version 8.2 of the MetUM. This code will be the basis for the chemistry and aerosol component of the new Earth-System model.

- **Chemical schemes** These are described below and are now held as seperate fortran modules. Details the model species and chemical equations are held in these modules. There are three main schemes using the ASAD system, two aerosol chemistry schemes, and three schemes retained which use a backward-Euler solver. *New for vn8.2: chemistry scheme suitable for both stratospheric and tropospheric chemistry including a parameterised isoprene scheme which is suitable for ozone prediction over the domain to 85 km. Also included are sulphur oxidation schemes suitable for the troposphere and stratosphere, and a scheme for the heterogenous chemistry of the troposphere.*

- **ASAD chemical solver system** This scheme allows integration of chemical schemes described in the fortran modules without pre-compilation. A Newton-

Raphson solver with error testing to adjust the iteration size is provided. *New for vn8.2: a routine which allows calculation of rate coefficients for aqueous-phase and tropospheric heterogenous reactions has been provided.*

- **Emissions** Emissions of species such as methane, carbon monoxide , NO$_x$ and sulphur dioxide are required in the model. These are input as monthly varying fields and are interpolated to the correct time and reformatted into the right units. The emissions are added to the lower boundary layer of the model. *New for vn8.2: Isoprene emissions are only emitted during the day, so the specified emissions are now adjusted to give a realistic diurnal variation. A small fraction of the* SO_2 *emissions are added as sulphate aerosol.*

- **Boundary conditions** The model tracers may be adjusted to the correct historical or scenario surface concentration through the lower boundary condition code. Top boundary conditions are applied for ozone and a few other species for tropospheric chemistry schemes. Finally, lateral boundary files are needed for limited area simulations. *New for vn8.2: The surface boundary conditions for methane have been extended to a greater range of species.*

- **Deposition schemes** Both wet and dry deposition schemes for chemical and aerosol species are provided. Two schemes for dry deposition are provided for chemical species with a simple scheme using specified dry deposition velocities or an interactive scheme which calculates both the aerodynamic and surface resistances interactively. *New for vn8.2: New species were added to the interactive deposition scheme to allow for the isoprene chemistry.*

- **Photolysis** Three schemes for photolysis are currently available: 1) a scheme based on lookup tables; 2) the Fast-J scheme initially installed for tropospheric chemistry; and 3) the FastJ-X scheme which is suitable for both tropospheric and stratospheric conditions. Once the FastJ-X scheme has been assessed for suitability in a variety of applications, the original Fast-J scheme will be withdrawn. *New for vn8.2: The FastJ-X scheme.*

- **MODE aerosol microphysics model** This model is described in detail by Mann et al. (2010), and is also described briefly below. *New for vn8.2: A boundary layer nucleation scheme has been added, and there have also been changes for stratospheric aerosol prediction.*

- **Coupling of trace gases to atmosphere model** The radiatively-active trace gases simulated in UKCA are coupled to the atmosphere model to affect radiative fluxes at every timestep.

- **RADAER scheme for direct aerosol forcing** The RADAER scheme calculates the aerosol optical depth and allows the simulated aerosol concentrations to affect the long-wave and short wave radiation.

- **Schemes to model Cloud Droplet Number Concentration (CDNC)** Two schemes for the prediction of CDNC are available: 1) A scheme using the parameterization of Jones et al. (2001) to convert from predicted cloud condensation nuclei; 2) A scheme based on Abdul-Razzal and Ghan (2000) which diagnoses

CDNC as a function of estimated cloud updraught velocity (Activate scheme). *New for vn8.2: Activate scheme for CDNC prediction.*

- **Unified Model Interface Panels** *New for vn8.2: A completely revized set of UMUI panels is being prepared for this release of UKCA.*
- **Documentation Paper** *New for vn8.2: A documentation paper to describe the technical aspects of UKCA is being prepared.*

4 Chemical schemes

The UKCA model has three basic chemistry schemes for tropospheric, stratospheric and combined (tropospheric plus stratospheric) conditions. These are integrated using a Newton-Raphson solver. In addition, aerosol chemistry schemes are included for tropospheric and stratospheric conditions. Three schemes which use a backward-Euler solver are also retained. Table 1 summarises the details of these schemes which us the Newton-Raphson solver.

Mechanism name	No. of species	No. of tracers	No. of reactions	solver
Tropospheric (Trop)	56	49	172	N-R
Stratospheric (Strat)	41	37	169	N-R
Strattrop (Strat-Trop)	75	72	282	N-R
Tropospheric aerosol	11	11	11	N-R
Stratospheric aerosol	12	12	23	N-R
Standard tropospheric	46	26	122	B-E
Standard trop + aerosol	73	33	130	B-E
Regional Air Quality	58	40	148	B-E

Table 1: Chemical schemes provided in the UKCA model. N-R: Uses Newton-Raphson chemical solver in the ASAD system. B-E: Uses explicit backward-Euler solver for each mechanism.

The stratospheric chemistry mechanism has been assessed on a multi-model framework (SPARC CCMVal, 2010) and the results are comparable with other models (see for example, Oman et al., 2010).

5 Photolysis

Earlier integrations of UKCA used a tabulated set of photolysis rates, however the Fastj on-line code was included and has been widely used for tropospheric chemistry. However this scheme is not suitable for stratospheric chemistry. Recently we have added the FastJ-X photolysis scheme that uses a wider range of wavelengths an is thus suitable for stratospheric chemistry.

6 UKCA-MODE Aerosol Scheme

The UKCA-MODE aerosol scheme uses a two-moment modal scheme where both the particle number density and the species mass are carried as prognostic tracers. The model framework follows that of the M7 model (Vignati et al., 2004), and table 2 shows the components and modes used by UKCA-MODE. The model contains a description of aerosol microphysics including nucleation of sulphuric acid, condensation, coagulation and cloud processing. Direct emissions of sulphate, black-carbon, and organic carbon are included, and secondary aerosol production from sulphur and terpene oxidation is taken into account. Aerosol removal by dry and wet deposition is also included. Dust and ammonium nitrate are not yet represented in the UKCA-MODE model, though these components are under development.

UKCA-MODE has been developed in an off-line chemical transport model TOMCAT, and has been ported to the MetUM to form a component of the UKCA model. This is described by Mann et al., 2010. Recent work (Mann et al., 2011) with this off-line model compares the results from the modal model with those of a detailed sectional model (Spracklen et al., 2005) running in the same chemical transport model. This comparison, after adjustment of parameters in the modal scheme, gave generally good agreement between the two models with the annual mean surface mass of sulphate, sea-salt, black carbon, and organic carbon agreeing to within 25% between the two schemes in nearly all regions. There were some differences: marine cloud concentration nuclei concentrations between 30^{o}S and 30^{o}N were sytematically higher in the modal scheme, and this was attributed to differences in size-resolved particle growth or cloud-processing.

UKCA-MODE is integrated with a tropospheric chemistry scheme which describes the inorganic chemistry of O_x-NO_x-HO_x-CO chemistry, together with near-explicit degradation schemes for methane, ethane, propane, and acetone. The chemical scheme also treats the degradation of the aerosol precursor species: sulphur dioxide (SO_2); dimethylsulphide (DMS); and monoterpenes. In addition, two tracers are used to represent species which are used in aerosol nucleation and deposition processes: sulphuric acid (H_2SO_4) produced from the oxidation of SO_2 with the hydroxyl radical (OH); and a secondary organic species representing the condensable species from monoterpene oxidation. A scheme for the oxidation of SO_2 within clouds by dissolved hydrogen peroxide (H_2O_2) and ozone (O_3) is also provided to give sulphate production rates which are used for in-cloud aerosol growth rates.

7 Aerosol chemistry

The chemistry for aerosol formation treats the decomposition of dimethylsuphide (DMS) and sulphur dioxide (SO_2) in the atmosphere. These are based on Pham et al. (1995), with revised rate coefficients. There is also a simple representation of mono-terpene oxidation. The model results have been compared against measurements, and for example, Figure 1 shows the comparison of model surface sulphate concentration against the IMPROVE network results. This shows the performance of two model versions: the standard model uses tabulated photolysis rates taken from a two-dimensional model, compared with a model version using the in-line fastJ-X pho-

Mode name	Soluble ?	Radius Range (μm)	Components
Nucleation	Yes	$\bar{r} < 0.005$	SO4, OC, OC2
Aitken	Yes	$0.005 < \bar{r} < 0.05$	SO4, SS, BC, OC, OC2
Accumulation	Yes	$0.05 < \bar{r} < 0.5$	SO4, SS, BC, OC, OC2, DU
Coarse	Yes	$\bar{r} > 0.5$	SO4, SS, BC, OC, OC2, DU
Aitken	No	$0.005 < \bar{r} < 0.05$	BC, OC
Accumulation	No	$0.05 < \bar{r} < 0.5$	DU
Coarse	No	$\bar{r} > 0.5$	DU

Table 2: MODE aerosol scheme, with component species: SO4, sulphate; SS, sea-salt; BC, Black Carbon; OC, Organic Carbon; OC2, Organic Carbon from biogenic VOCs; DU, dust.

tolysis scheme. The in-line scheme is clearly expected to be better than the off-line as it can respond to variations in the model ozone and cloud profiles. As can be seen the fastJ-X version does show improvement in the comparison with measured sulphate concentrations.

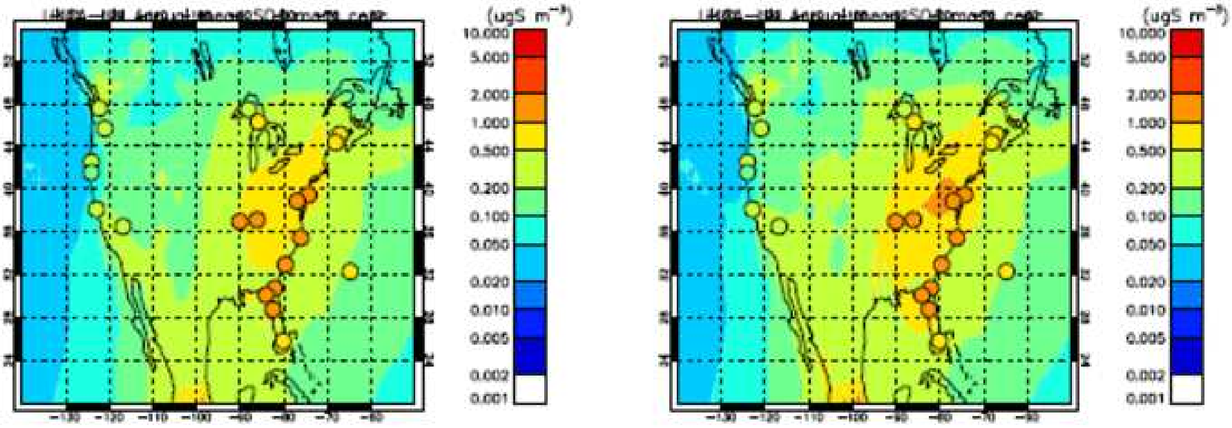


Figure 1: Comparison between annual mean surface sulphate aerosol concentrations and measurements from the IMPROVE network. Left: UKCA using a table of photolysis rates; Right: UKCA using the fastJ-X photolysis model.

The budget of SO_2 production and loss in the UKCA model is compared with those from other models in table 3. UKCA has the highest production of SO_2 from DMS, and slightly lower cloud oxidation rates than the other two models.

8 Assessment of aerosol modelling performance against observations.

Two HadGEM simulations have been run using nudged meteorology and monthly aerosol precursor and primary emissions for the period 2000-2006. The difference be-

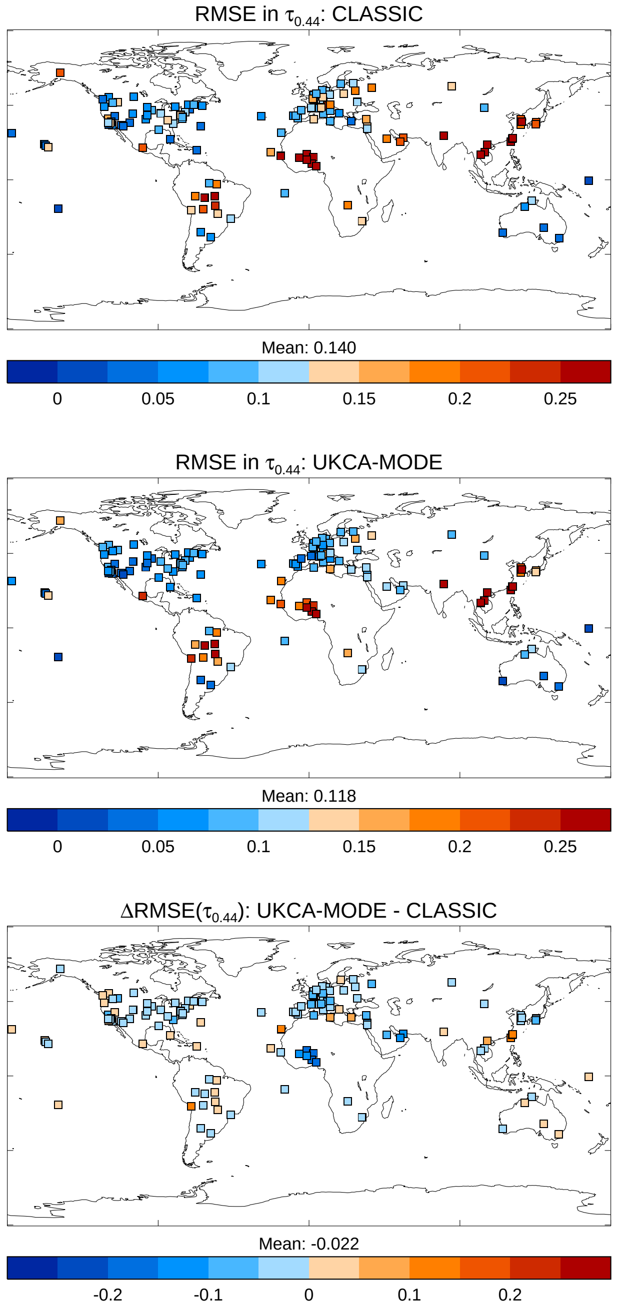
Model or author	UKCA	LMDZ	Chin et al.
Base year	2000	2000	1985
Industrial Emission	61.8	63.0	67.4
Natural Emission	12.4	7.8	6.7
DMS + OH $\Rightarrow$ SO_2	16.3	11.3	
DMS + NO_3 $\Rightarrow$ SO_2	7.9	5.2	
Total photochemical source			21.5
Total Sources	98.3	91.9	95.6
SO_2 + OH	14.5	14.0	7.5
HSO_3 + H_2O_2 (Aq)	24.2	27.9	
SO_3 + O_3 (Aq)	13.0	21.1	
Total In-cloud oxidation			41.6
Dry deposition	31.4	22.7	26.6
Wet deposition	16.0	6.2	19.9

Table 3: Annual SO_2 fluxes in UKCA and the LMDZ model (Boucher et al., 2002) for the year 2000, and taken from Chin et al. (1996). Units are Tg [S]/year. The total sources for the LMDZ model includes a contribution of 4.6 Tg/year from oxidation of H_2S and DMSO.

tween the simulations is the aerosol module: the first simulation uses CLASSIC aerosols while the second uses UKCA-MODE aerosols. Monthly total aerosol optical depths are then compared against sun-photometer measurements at 134 AERONET (See Holben et al., 2001) sites worldwide by computing root-mean square errors (RMSE), correlations, and biases between modelled and observed timeseries. The selected AERONET sites provide at least 24 valid monthly means over the simulated period (data version 2, quality level 2). Figure 2 shows the distribution of RMSE against AERONET for CLASSIC (top panel), UKCA-MODE (middle panel), and their difference (bottom panel). Each coloured square represents a good-quality AERONET measurement site. The two aerosol modules share common patterns: RMSE are lower over Europe, North America, and Australia, and larger over South America, Africa, and Asia. However, using UKCA-MODE decreases the errors at almost all sites, with larger improvements in sub-saharan Africa. On a global average, RMSE decreases by 0.02 (16 %) when UKCA-MODE is used. UKCA-MODE also improves correlation and decreases bias against AERONET (not shown).

9 Aerosol radiative forcing in CLASSIC and UKCA-MODE.

Aerosols emitted into the atmosphere by human activities between 1860 and 2000 exert a radiative forcing of the climate system. The forcing is due to the increase in the amount of solar and terrestrial radiation that is scattered and absorbed in the atmosphere, termed direct forcing. The forcing is also due to the increase in the amount of cloud condensation nuclei that leads to the formation of more, smaller cloud droplets. This is the first indirect forcing. On a global average, aerosol forcing is


RMSE in $\tau_{0.44}$: CLASSIC

Mean: 0.140

RMSE in $\tau_{0.44}$: UKCA-MODE

Mean: 0.118

ΔRMSE($\tau_{0.44}$): UKCA-MODE - CLASSIC

Mean: -0.022

Figure 2: Root-mean square error between observed timeseries of total aerosol optical depth at 0.55 μm at 134 AERONET sites, represented by coloured squares, for (top) the CLASSIC aerosol module, (middle) the UKCA-MODE aerosol module, and (bottom) their differences.

negative, leading to a cooling of the climate system that counteracts the greenhouse gas warming.

Direct and first indirect forcing have been quantified in HadGEM simulations using CLASSIC or UKCA-MODE, with the same meteorology and the same aerosol precursor and primary emissions: the only difference is the aerosol module. The top row of Figure 3 shows the change in aerosol aerosol optical depth between 2000 and 1860, called anthropogenic aerosol optical depth, as obtained using CLASSIC (left column) or UKCA-MODE (middle column), and their difference (right column). Differences in aerosol residence times between CLASSIC and UKCA-MODE yield larger anthropogenic optical depths in UKCA-MODE on a global average. Anthropogenic optical depths are larger in UKCA-MODE over the high latitudes of the north hemisphere. This is partially compensated by smaller anthropogenic optical depth in biomass burning areas of Africa, South America and Indonesia, and in industrial China.

Figure 3 also shows the radiative forcing exerted by anthropogenic aerosols in cloud-free sky (middle row) and all-sky (bottom row) conditions. In cloud-free sky, only the direct forcing is active. It increases from -0.6 Wm^{-2} in CLASSIC to -1.0 Wm^{-2} in UKCA-MODE. This increase by a factor 1.7 is simply due to the commensurate increase in anthropogenic aerosol optical depth between the two aerosol modules. In all-sky, where both direct and first indirect forcings are active, both aerosol modules produce the same forcing of -1.7 Wm^{-2}. However, this agreement on the global average masks important regional differences. Over the maritime stratocumulus regions off the shore of Namibia, Peru, and California, UKCA-MODE aerosols exert weaker (less negative) forcings. This is a consequence of the key difference between CLASSIC and UKCA-MODE, and a strength of UKCA-MODE. In CLASSIC, increasing aerosol mass automatically increases the number of aerosols and cloud condensation nuclei. In UKCA-MODE, increasing aerosol mass may not increase aerosol number, as aerosols may mix internally into the same particle: that process has the ability to dampen cloud susceptibility to aerosol changes and produce weaker first indirect aerosol forcing, as shown in Figure 3. In addition, weaker forcings obtained by UKCA-MODE over the Congo Basin and the Namibian stratocumulus deck are also a consequence of both a smaller anthropogenic optical depths there, and a greater absorption by the biomass-burning aerosols that overlie clouds. Over north hemisphere continents, UKCA-MODE typically produces a stronger (more negative) aerosol forcing. This is due to increased anthropogenic optical depths in UKCA-MODE. In China, the increased first indirect aerosol forcing produced by UKCA-MODE is certainly due to coated black-carbon aerosols, an important process that is not represented in CLASSIC.

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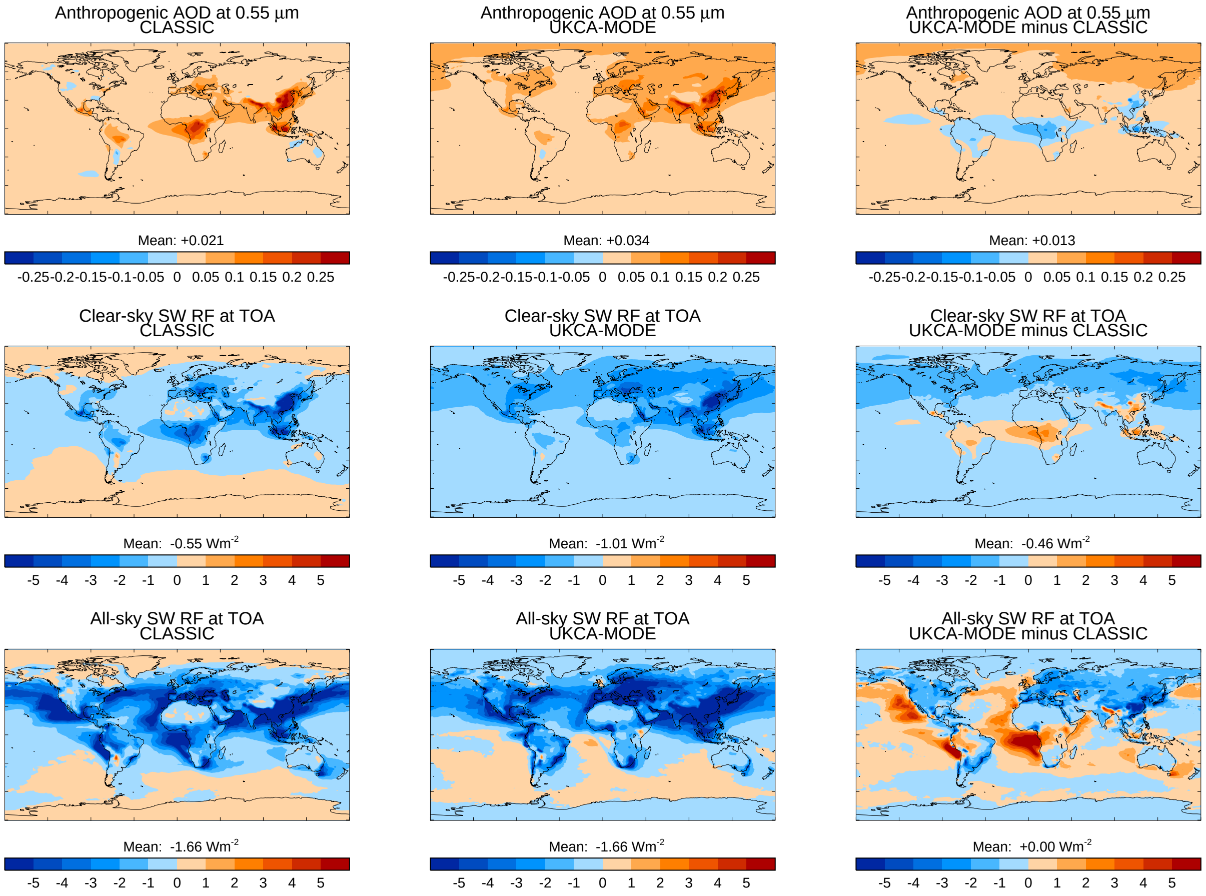

Figure 3: Top row: Anthropogenic aerosol optical depth at 0.55 μm in CLASSIC (left) and UKCA-MODE (middle) and their difference (right). Middle row: Same but for the clear-sky aerosol radiative forcing at the top of the atmosphere, in Wm^{-2}. Bottom row: Same but for the all-sky aerosol radiative forcing at the top of the atmosphere, in Wm^{-2}.

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