Achieving an improved sulphur cycle in the HadGEM model through on-line calculation of oxidant concentrations

Jamie G.L. Rae, Colin E. Johnson and Fiona M. O'Connor

UKCA is a new chemistry and aerosol model which is being developed and coupled to the Met Office Unified Model (UM) as part of the Hadley Centre HadGEM climate models. Concentrations of oxidants generated at each model timestep by the tropospheric chemistry component of UKCA can now be used to drive the UM sulphur-cycle scheme, which previously used prescribed monthly-mean oxidant concentrations calculated off-line. This has the advantage that the dependence of oxidant concentrations on meteorological variables is now explicitly represented. Here, we present results for the global and zonal effects of the introduction of this coupling; we also validate the model results with surface and satellite observations.

1.Model description

- HadGEM2-A (atmosphere-only).
- N96 horizontal resolution (1.875º in longitude,1.25º in latitude).
- 38 vertical levels.
- Sulphur-cycle scheme:
  - Based on that in HadGAM1 (Jones et al., 2001).
  - Improvements described by Bellouin et al. (2007) and Rae et al. (2007).
  - Aqueous S(IV)+O_3 reaction included for the first time.
  - SO_2 emissions appropriate for the 1990s.
- UKCA tropospheric chemistry scheme:
  - 26 advected tracers; 20 non-advected species.
  - Emissions (at surface and higher levels).
  - 88 bimolecular, 14 termolecular and 20 photolysis reactions.
  - Wet and dry deposition.
- Coupling introduced:
  - Oxidant concentrations from UKCA now used in HadGEM sulphur cycle.
  - One-way coupling (no feedback to UKCA).

2. Model experiments

Five-year experiments:
- CTRL: Standard prescribed monthly-mean oxidants.
- UKCA_OL: On-line oxidant concentrations from UKCA.
- UKCA_PR: Prescribed monthly-mean oxidants from UKCA output in UKCA_OL.

- Largest difference is between CTRL and UKCA_OL.

→ Source of oxidant fields more important than whether on-line or prescribed.

- ~50% difference in H_2O_2 consistent with ~25% difference in HO_2 (not shown).

Table 1: 5-year mean tropospheric oxidant burdens in CTRL, and differences between experiments. (*) denotes statistical significance at 95% level.

Species	CTRL (Tg)	UKCA_OL—CTRL (%)	UKCA_OL—UKCA_PR (%)
OH*	1.75×10^{-4}	–8.1 (*)	–0.1
H_2O_2	3,81	–43.1 (*)	0.0
O_3	6.87×10^{2}	–47.4 (*)	0.0

* Adjusted for daylight fraction

3. Effect on reaction budgets

Table 2: 5-year mean tropospheric reaction budgets in CTRL, and differences between experiments. (*) denotes statistical significance at 95% level.

Reaction	CTRL (Tg[S]/yr)	UKCA_OL—CTRL (%)	UKCA_OL—UKCA_PR (%)
SO_2+OH	16.0	–7.4 (*)	–0.1
S(IV)+H_2O_2	16.6	–16.8 (*)	+12.2 (*)
S(IV)+O_3	24.2	+17.2 (*)	–7.4 (*)

- Zonal mean reaction budgets were studied for January and July.
- In most cases, differences between zonal mean reaction budgets in different experiments are within one standard deviation of zero.
- In Figure 1, error bars showing one standard deviation are included where this is not the case.
- In other cases, no conclusion can be reached about significance of zonal mean results.
- Aerosol-climate feedbacks may be affecting results.
- Runs without such feedbacks now being performed to ascertain significance of results.

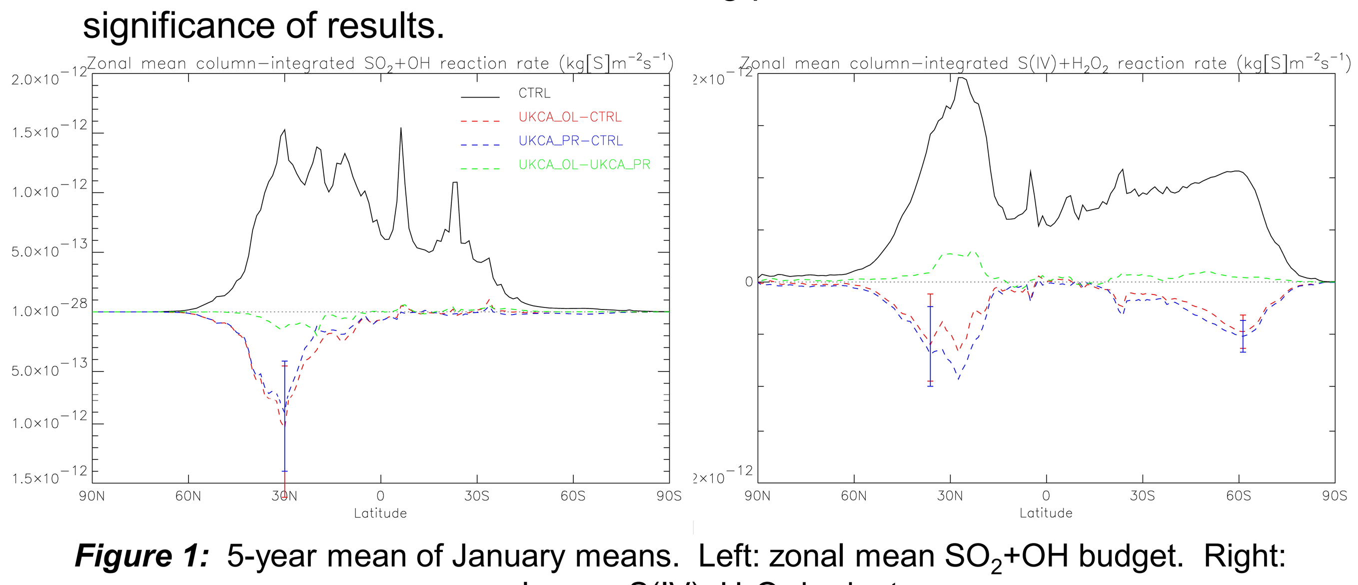


Figure 1: 5-year mean of January means. Left: zonal mean SO_2+OH budget. Right: zonal mean S(IV)+H_2O_2 budget.

4. Effect on sulphate aerosol

- In Table 3, in most cases differences in tropospheric burdens are not statistically significant.
- Zonal means were also studied.
  - In all cases, differences in zonal means were within one standard deviation of zero.
  - Again, runs without aerosol-climate feedbacks will help to ascertain significance of zonal mean results.

Table 3: 5-year mean tropospheric sulphate burdens in experiment CTRL, and differences between experiments. (*) denotes statistical significance at 95% level.

Mode	CTRL (Tg[S])	UKCA_OL—CTRL (%)	UKCA_OL—UKCA_PR (%)
Aitken	0.092	+2.4 (*)	–1.7 (*)
Accumulation	0.422	–0.7	–0.2
Dissolved	0.026	+0.2	–0.8
TOTAL	0.540	–0.1	–0.5

5. Validation against observations

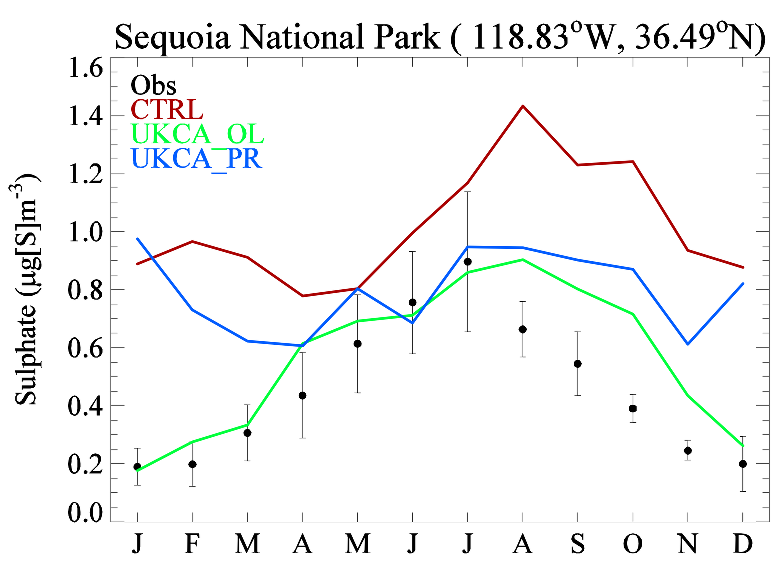
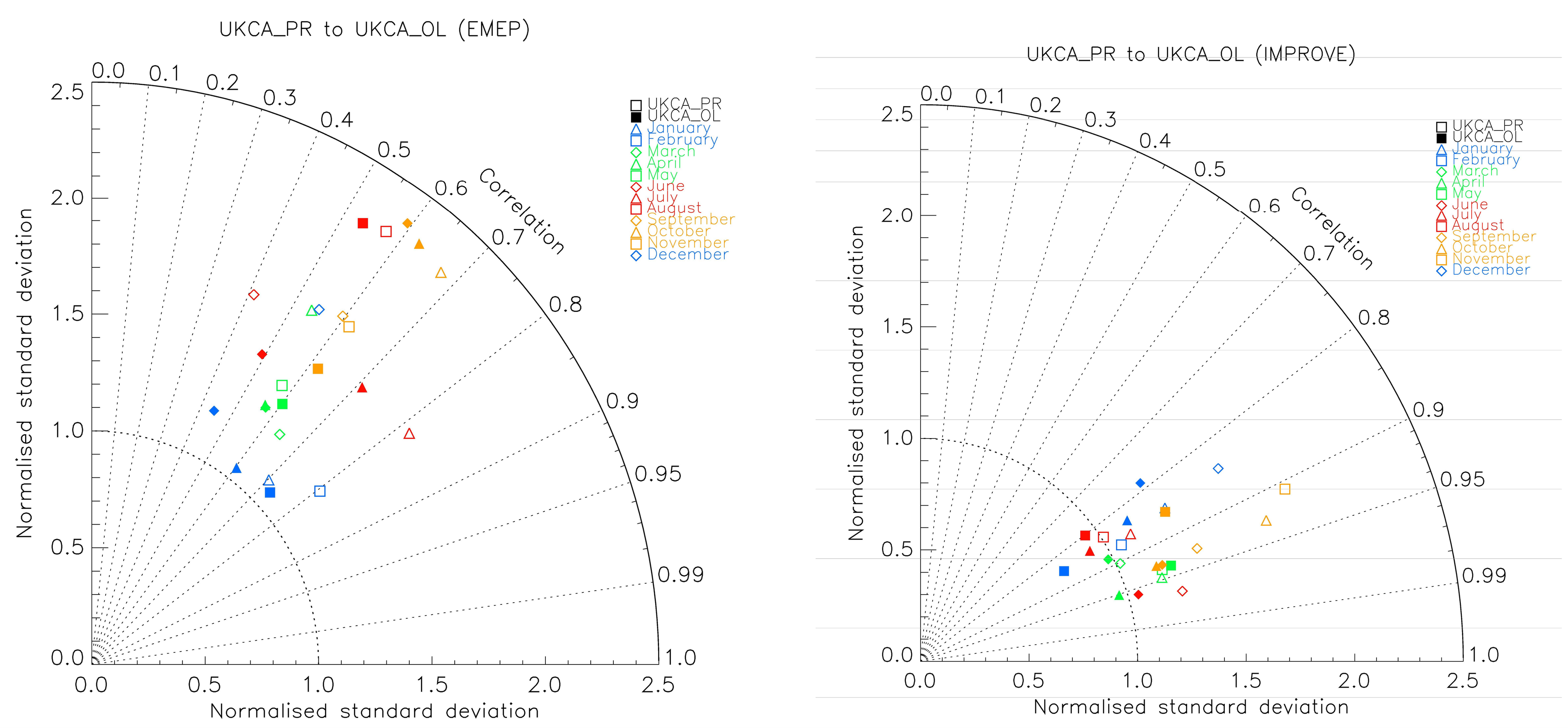


Figure 2: Annual cycle in sulphate concentrations at an observing site in the united states. Black circles: observations. Coloured lines: model output.

- Figure 2 shows the annual cycle in sulphate concentration at an observing site in the United States.
- At this site, the advantages of using on-line oxidants are obvious.
- Improvement seen at some other sites, but not all.
- Figure 3 shows Taylor plots (see Taylor, 2001) comparing modelled and observed sulphate concentrations.
- Correlation between model and observations is shown, as is normalised standard deviation (std. dev. in model output / std. dev. in observations).
- This is done for European EMEP observations (see **www.emep.int**) and North American IMPROVE observations (see **http://vista.cira.colostate.edu/improve**).
- Comparison is done for UKCA_OL and UKCA_PR, to allow comparison between prescribed and online oxidants from same source.
- No general improvement or deterioration in comparison between model and observations when we move from UKCA_PR to UKCA_OL – there is improvement in some months, and deterioration in others.

Figure 3: Taylor plots comparing modelled sulphate concentrations with observations. Left: EMEP observations (Europe). Right: IMPROVE observations (North America). Open symbols: UKCA_PR. Closed symbols: UKCA_OL.



Discussion

Overall, little apparent improvement was observed in model output, but this modification to the model is useful for several reasons:

- Prescribed oxidants not available for full 21st century runs. Can now use on-line oxidants in this case.
- Feedback from meteorology on oxidants may be important at higher spatial and temporal resolution, so on-line oxidants useful for forecasting air quality.
- Can now investigate climate-chemistry-aerosol feedbacks.
- Oxidants now calculated at each timestep, so can validate against instantaneous field measurements.
- Can implement additional aerosol schemes (e.g. nitrate) more easily.

It is a positive development that this additional functionality has been introduced without any negative impact on model output.

References

Bellouin N., et al. (2007), Hadley Centre Technical Note No. 73 (available at www.metoffice.gov.uk/research/hadleycentre/pubs/HCTN/index).

Jones A., et al. (2001), *J.Geophys.Res.,* 106, 20293—20310.

Rae J.G.L., et al. (2007), *J.Geophys.Res.,* 112, doi:10.1029/2006JD007826.

Taylor K.E. (2001), *J.Geophys.Res.*, **106**, D7, 7183—7192.

Met Office Hadley Centre, FitzRoy Road, Exeter, Devon, EX1 3PB, United Kingdom
Tel: +44 (0)1392 884007 Fax: +44 (0)1392 885681
Email: jamie.rae@metoffice.gov.uk