Challenging UKESM1 with SO2 and sulphate observational data to evaluate the aerosol sulphur cycle

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UKESM1 is the latest generation Earth system model to be developed in the UK. It simulates the core physical and dynamical processes of land, atmosphere, ocean and sea ice systems which are extended to incorporate key marine and terrestrial biogeochemical cycles. These include the carbon and nitrogen cycles and interactive stratosphere-troposphere trace gas chemistry. Feedbacks between these components that have an important amplifying or dampening effect on the physical climate, and/or change themselves in response to changes in the physical climate are also included. One focus for the future development of UKESM1 is improved treatment of sulphur processes, including emission, chemical processing and deposition in the aerosol-chemistry scheme, UKCA-Mode. These processes span land-atmosphere and ocean-atmosphere boundaries and can therefore impact feedbacks between these systems. Emissions of SO2 can be oxidised to form sulphate aerosol, which plays a key role in both acid deposition, atmospheric aerosol loading and cloud properties, thereby directly contributing to the Earth’s radiative balance. Good representation of sulphur processes in UKESM1 is therefore essential for constraining uncertainties associated with the impacts of aerosols on the Earth system and thus understanding the global climate. Here we challenge UKESM1 with observations of SO2 and sulphate from ground-based measurement networks in Europe and the USA, and of SO2 from the Ozone Monitoring Instrument (OMI). We use these to evaluate temporal and spatial biases in the model's simulation of SO2 and sulphate.

We find that UKESM1 captures the historical trend for decreasing concentrations of atmospheric SO2 and sulphate in both Europe and the USA over the period 1987 to 2014. However, in the polluted regions of the Eastern USA and Europe, UKESM1 over-predicts surface SO2 concentrations by an average of 320-340%, while under-predicting surface sulphate concentrations by 25-35%. In the cleaner Western USA, the model over-predicts both surface SO2 and sulphate concentrations by 1200% and 150% respectively. The variability in
the direction of UKESM1’s bias according to species and region suggests that the model bias may be driven differently depending on species and region. These drivers likely result from uncertainty in aspects of the sulphur cycle, including SO2 emission, loss processes (oxidation and deposition) or transport. To evaluate UKESM1 at the global scale we use a newly available data product for total column SO2 (TCSO2) from OMI. We find that UKESM1 over-predicts TCSO2 over much of the globe, particularly the large source regions of India, China, the USA and Europe as well as over background regions, including much of the ocean.

In this study we also assess changes to UKESM1’s SO2 dry deposition parameterization. These changes increase SO2 dry deposition to land and ocean surfaces, thus reducing atmospheric SO2 and sulphate concentrations, and ultimately reducing cold bias in UKESM1’s simulation of mid 20th C global mean surface temperatures. In comparison with the ground based and satellite observations, we find that the changes reduce UKESM1’s over prediction of surface SO2 concentrations and TCSO2.