Seasonal Variability of Historical Sulfate Aerosol Formation in Earth System Model with Interactive Chemistry

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Abstract

Over the historical period, emissions of greenhouse gases and short-lived climate forcers (SLFCs) including aerosol precursors, O_3 precursors and CH_4 have broadly increased. We ask how seasonal variability of SLFCs controls aerosol and cloud radiative forcing and aim to answer this using Earth system modelling. This is important because a lack of understanding of heterogeneous aerosol-climate effects is hampering our understanding of historical climate change. Using the UK Earth System Model 1 (UKESM1), we investigate how sulfate aerosols form under emission and oxidant changes between 1850 and 2014. We analyse simulation output from the Aerosol Chemistry Model Intercomparison Project (AerChemMIP) atmosphere-only transient experiment designed to isolate their effects on the Earth system responses. We show that emission timing determines oxidation tendency via the available oxidant and meteorological properties such as clouds. In the UKESM1, SO_2 reacts with OH in the gas phase and O_3 and H_2O_2 in the aqueous phase. Up to 80% of total sulfate production is via gas phase oxidation in boreal summer when high OH and low cloud cover are observed. The opposite is true for wintertime when aqueous phase reactions with O3 and H_2O_2 form up to 90% of aerosol. This work shows that the same amount of SO_2 emitted at different times does not form aerosol with the same aerosol size distribution, contributing to changes in radiative effects at different times of the year. We present an analysis of these monthly changes of oxidants and emissions to sulfur oxidation, aerosol and cloud properties and connect these to effective radiative forcing. Ultimately, this work contributes to improving our process-level understanding of Earth system models that interactively simulate aerosol from precursors and aims to improve the accuracy of aerosol radiative forcing predictions.

Early Career Scientist

YES, I am an early career scientist.

IGAC Activities

CCMi: Chemistry Climate Model Initiative