How do atmospheric oxidants change aerosol climatic impacts?

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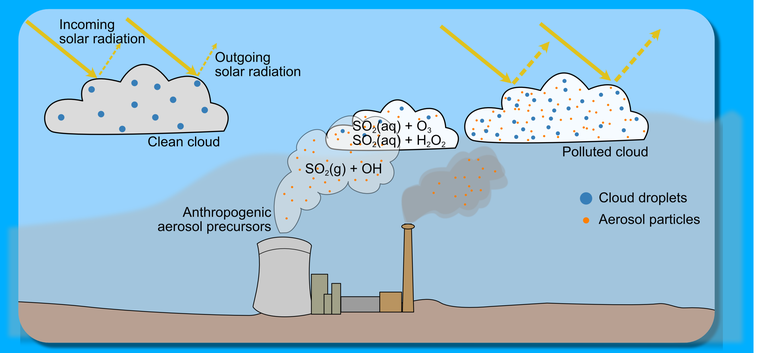

In a nutshell

Sulfate aerosols play a significant role in shaping the climate by inducing cooling by directly reflecting sunlight and also by modifying cloud properties. Sulfur dioxide (SO_2) is an important sulfate aerosol precursor with the largest sources coming from anthropogenic activity. The oxidants that SO_2 react with are controlled by complex chemical processes.

In this work, we analysed the output from the UK's Earth System Model1 from oxidation processes to radiative effect to tease out the effects of oxidants on aerosol and climate.

We show how ❶ OH, O_3 and H_2O_2, which are the main SO_2 oxidants response to O_3 precursors and CH_4 increases. ❷ SO_2 oxidation follows SO_2 emissions but is perturbed by oxidant changes. SO_2 + OH is the dominant oxidizing process and is also the most sensitive to OH changes.

❸ Historical changes in aerosol size distribution is affected by oxidant changes. ❹ Ultimately, historical changes in O_3 precursors affect the aerosol abundance in the nucleation mode and have a cooling effect through scattering but CH_4 has a warming effect by reducing aerosol cloud interaction.

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Simulation design

Existing UKESM1 outputs from the AerChemMIP were used in this work. All experiments have prescribed sea-surface temperature (SST). We gratefully acknowledged the UKESM1 AerChemMIP team for provision of the data.

Table 1. List of AerChemMIP simulations used. "Hist" means historical values and "1850" means the respective emission is set to 1850 throughout the runs.

Experiment ID	CH_4	Aerosol precursors	Ozone precursors	Purpose
histSST	Hist	Hist	Hist	Historical
histSST-piAer	Hist	1850	Hist	Target SO_2
histSST-piO3	Hist	Hist	1850	Target O_3
histSST-piCH4	1850	Hist	Hist	Target OH

❶ Oxidants

In the UKESM1 model, SO_2 reacts with OH in the gas phase, and O_3 and H_2O_2 in the aqueous phase to form sulfate aerosols. AerChemMIP designs the experiment such that oxidant precursor effects can be isolated. Here we show the four important chemical species related to sulfate aerosol formation.

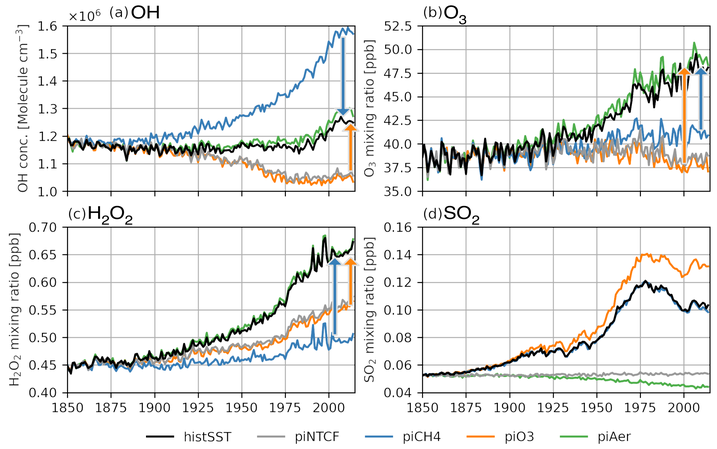


Figure 1. Global average OH concentration, O_3 mixinng ratio, H_2O_2 mixing ratio and SO_2 mixing ratio from experiments described in Table 1.

❷ Sulfur oxidation tendency

Changes in oxidant level affect sulfur budget terms by modifying the oxidation tendency. The SO_2-OH channel is the most sensitive to both O_3 precursor and CH_4 concentration.

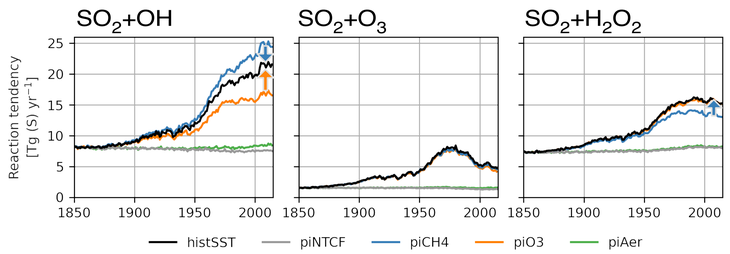


Figure 2. Global sulfur dioxide reaction tendencies

❸ Aerosol and cloud properties

Changes in reaction tendencies in ❷ have a knock-on effect on aerosol formation. Less OH in piO3 lowers aerosol in the nucleation mode and aerosol optical depth. Cloud droplet number concentration is also perturbed by oxidant changes, with historical CH_4 decreasing CDNC globally while O_3 precursors increases CDNC.

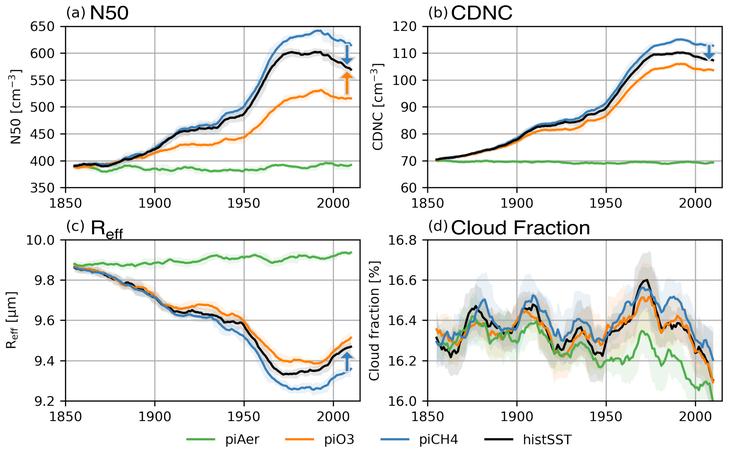


Figure 3. Global decadal mean aerosol and cloud properties at 1 km above ground.

❹ Radiative forcing

Effective radiative forcing (ERF) is decomposed into aerosol instantaneous radiative forcing (IRF), ERF from clean and clear sky ($ERF_{cs,clean}$) and cloud radiative effect (CRE).

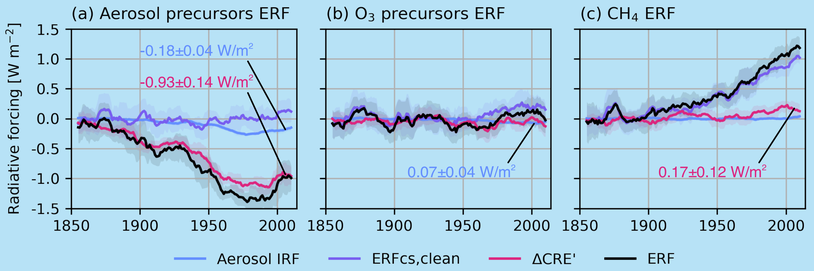
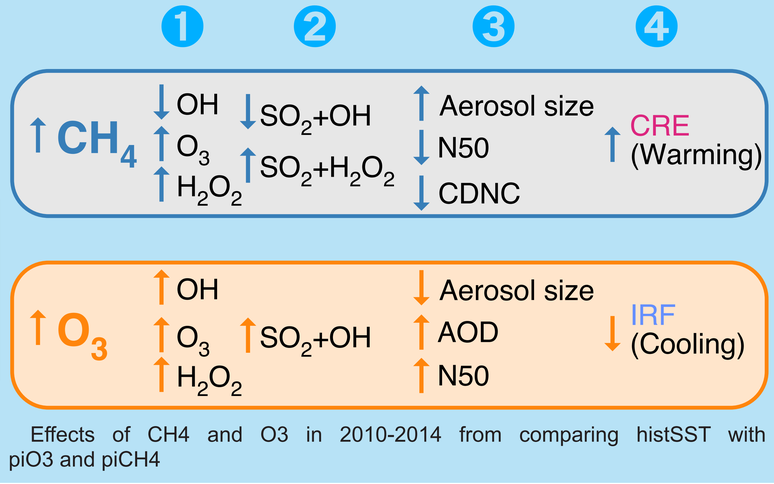


Figure 4. Aerosol effective radiative forcing (ERF) due to aerosol precursors, O3 precursors and CH4



Effects of CH4 and O3 in 2010-2014 from comparing histSST with piO3 and piCH4