How do historical changes in oxidants affect sulfate aerosol formation in CMIP6 simulations?

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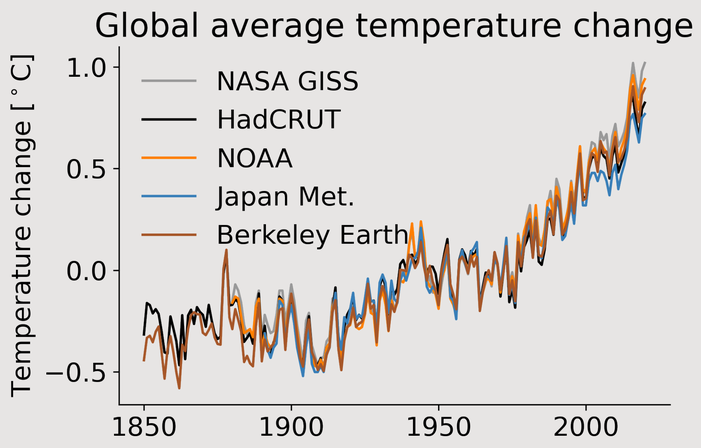
In a Nutshell



Historical observations show a pause in the growth of global mean surface temperature in 1950-1980, a period when SO_2 emissions were increasing rapidly.

This raises the question as to what extent sulfate aerosol affect global mean surface temperature.

Globally, SO_2 emissions peaked around 1980 but continue to increase in some regions. Additionally, the location of emissions is shifting equatorward.

The lifetime of SO_2 is short (2 days) meaning its impact is localised near to source.

We conducted an analysis of SO_2 oxidation pathways from UKESM1 CMIP6 histSST experiments to explain changes in sulfate aerosol formation.

Our results show that SO_2 oxidation trends follow SO_2 emission which peaked in Europe in 1980 whereas in Eastern Asia, oxidation continues to increase.

We found that SO_2 + OH is the dominant oxidizing process and is also the most sensitive to oxidant changes.

By reducing OH, the effect of historical methane emissions has been to increase oxidation in the gas-phase with implications for both aerosol and cloud radiative forcing.

Historical changes in O_3 precursor emission increased SO_2 oxidation by 30%

Introduction

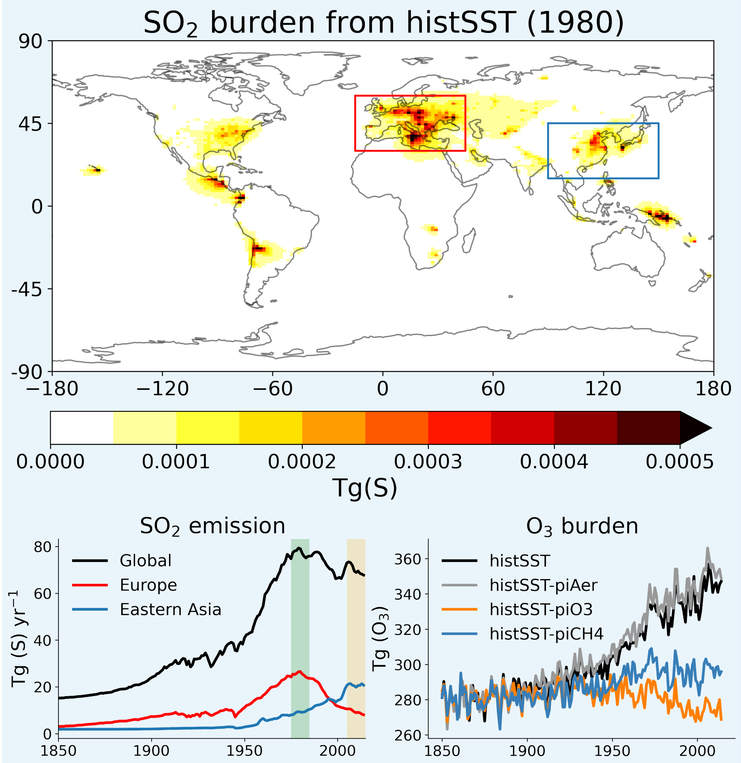


Figure 1. (Top) Annual average tropospheric SO_2 burden from histSST experiment. (Left) Annual average SO_2 emission from histSST experiment. (Right) Annual average O_3 burden from 4 experiments

Emissions of SO_2 offset some of warming by CO_2 and CH_4 through their formation of aerosols, which modify the properties of clouds.

SO_2 emission location is shifting away from the European region to Eastern and Southern Asia in the past 30 years.

This work studies the sensitivity of aerosol formation to oxidant changes in Earth system models.

Methods

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
histSST	Hist	Hist	Hist
histSST-piAer	Hist	1850	Hist
histSST-piO3	Hist	Hist	1850
histSST-piCH4	1850	Hist	Hist

Results

SO_2 and oxidant burdens and reaction tendencies

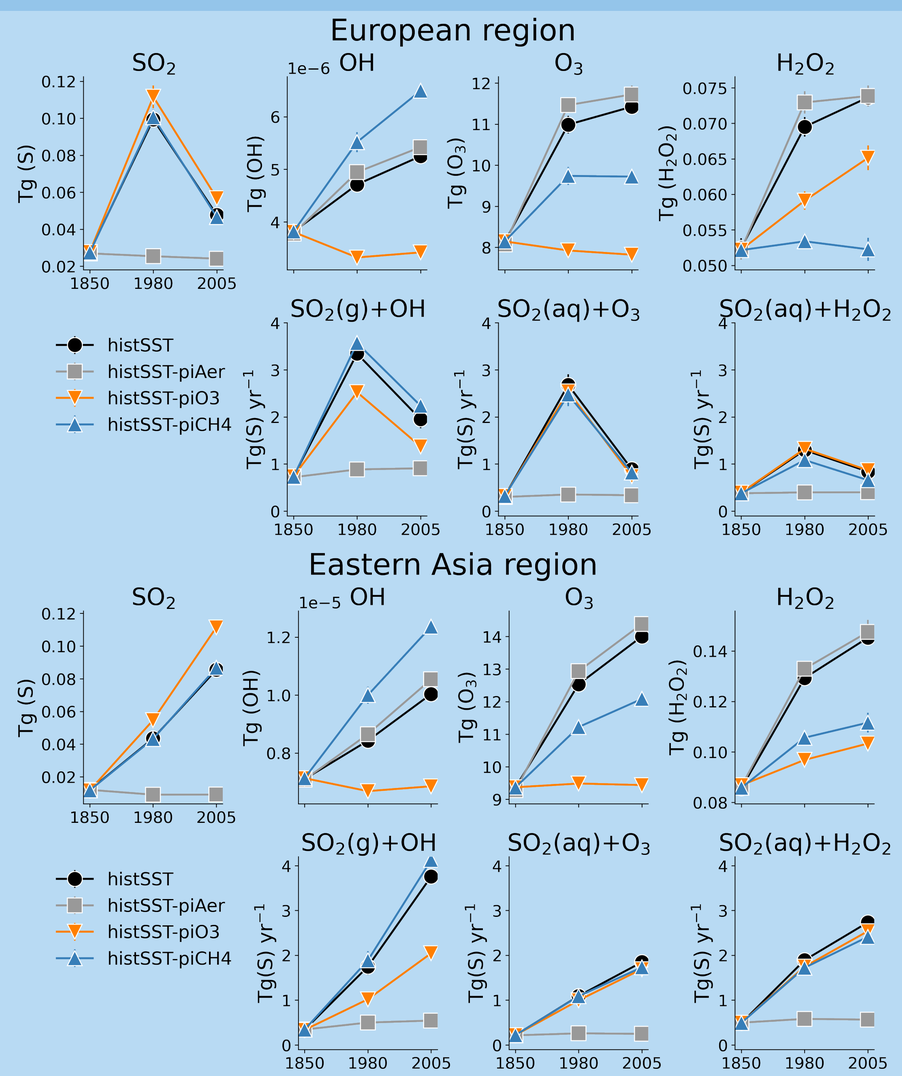


Figure 2. Annual average of SO_2, OH, O_3 and H_2O_2 burdens and respective reaction mass tendencies for European region and Eastern Asia region, calculated for 0-10 km above ground.

Key Points

1. SO_2+OH is the dominant oxidation pathway for both regions. SO_2+H_2O_2 is the smallest SO_2 sink in the European region, while, in Eastern Asia, O_3 and H_2O_2 channels are equally important.

2. Since CH_4 acts as OH sink, the excess OH in hisSST-piCH4 reacts with SO_2 that would have reacted with H_2O_2. These two reactions compensate each other, resulted in unchanged SO_2 burden compared to histSST.

3. At peak emission in European region, 1980, aerosol and O_3 precursors are accountable for 4.5 and 0.8 Tg(S) yr^{-1} of SO_2 oxidation of total 7.5 Tg (S) yr^{-1} oxidation, respectively.

4. Reduced O_3 production in histSST-piO3 does not affect SO_2+O_3 tendency, but reduces SO_2+OH tendency by 30% and 50% in European and Eastern Asia, respectively.

Future Plans

We show changes to SO_2 oxidation across the historical period 1850-2014 in terms of reaction pathway, tendency and branching ratio between the three channels included in UKESM1. We will go on to study

How did sulfate aerosol evolve across this period?

How did cloud properties change?

How do these changes connect to radiative forcing?

Furthermore, can we attribute the effect of individual emissions changes on aerosol formation?

Ultimately, we seek to quantify the underpinning aerosol production channels, sulfate burden, and important radiative effects, which will drive climate responses.

We will extend this study to other CMIP6 models.

References

Mulcahy, J.P. *et al.* (2020) 'Description and evaluation of aerosol in UKESM1 and HadGEM3-GC3.1 CMIP6 historical simulations', *Geoscientific Model Development,* 13(12), pp. 6383–6423. Available at: https://doi.org/10.5194/gmd-13-6383-2020.

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