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# Strong impacts on aerosol indirect effects from historical oxidant changes

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The contribution by aerosol-cloud interactions to the total preindustrial to present-day anthropogenic forcing is highly uncertain. Previous studies have pointed out natural background emissions and parameterizations of the microphysics as large contributors to this uncertainty. Here we highlight the oxidant concentrations as a third important factor.

Aerosols are not only emitted directly into the atmosphere, but also formed in situ through nucleation from condensable gases. The oxidation process leads to gases with lower saturation vapor pressure, increasing the ability to form aerosols under atmospheric conditions. As the oxidation capacity of the atmosphere has changed since preindustrial times due to anthropogenic activity, so have the atmospheric conditions for secondary aerosol formation. Previous model studies of the preindustrial to present-day aerosol indirect effect usually applied oxidants from present-day when performing model simulations with present-day as well as preindustrial aerosols and aerosol precursor gases.

Here, we show that if we instead expose the precursor gases to oxidants of their era, the total preindustrial to present-day aerosol indirect effect changes from -1.32 $Wm^{-2}$ to -1.07 $Wm^{-2}$. Exposing the preindustrial precursor gases to an atmosphere with weaker oxidation capacity surprisingly results in increased aerosol formation, increased cloud droplet number concentrations and a brightening of the clouds. This happens because the reduced oxidative power of the atmosphere extends the lifetime of the precursor gases, allowing them to be transported higher up in the atmosphere and horizontally towards more remote areas where the coagulation sink is low and the susceptibility of the cloud albedo to aerosol changes is high. The importance of different chemical reactions is shifted by the oxidation changes, resulting in more production of condensate, giving larger aerosols which more easily activate as cloud condensation nuclei. The historical change of the concentration of the nitrate radical is shown to be the most important oxidant change from preindustrial to present-day. The model treatment of secondary aerosol formation from marine emissions are highlighted as important for the magnitude of the result.

The findings of this study opens for a discussion of a common way to treat the oxidants when modeling the preindustrial to present-day aerosol indirect effect, as well as other aerosol effective radiative forcings.