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Long-term trends in oxidant budgets of the northern hemisphere constrained by alkanes and alkyl nitrate ratios

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Our study aims to understand how the concentrations of ozone (O_3) and hydroxyl radicals (OH), and as such the tropospheric oxidizing capacity, have changed since the mid-20th century, largely as a result of anthropogenic emissions of air pollutants. We do this through a combination of field observations and numerical simulations. Global emissions of O_3 precursors, such as volatile organic compounds (VOCs), oxides of nitrogen (NO_x) and carbon monoxide (CO), have changed substantially since preindustrial times, resulting in changes to the tropospheric budgets of O_3 and OH. However, the absence of long-term observational records of HO_x and NO_x results in large uncertainties associated with the current understanding of these past changes.

We use measured long-term variations of alkane and alkyl nitrate concentrations from firn air to constrain global atmospheric model scenarios, because the formation of nitrates from alkanes is closely linked to chemistry involving HO_x and NO_x. This enables us to assess changes in the O_3 and OH radical budgets of the northern hemisphere troposphere. We have used the UK community global chemistry-climate model (UKCA) in its current configuration as part of the UK community Earth System Model (UKESM-1) to carry out transient model experiments from 1960 to the present day. We use global emissions data from the CMIP5 and also from the forthcoming CMIP6 model intercomparison projects. CMIP6 emissions show significantly higher NO_x and C3-C5 alkane fluxes compared to CMIP5 resulting in increased nitrate formation. The model is validated with surface and airborne observations of O_3 precursors, alkanes and alkyl nitrates, as well as with long-term trends for alkanes and nitrates obtained from firn air in Greenland.