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Indirect effect of changing aerosol concentrations on methane and ozone radiative forcing

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Atmospheric aerosols interact with climate in number of complex ways and quantifying the overall effect remains the dominant uncertainty in estimating anthropogenic climate forcing (IPCC, 2013). The radiative forcing (RF) caused by the direct effect of aerosol interacting with radiation is estimated at -0.35 (-0.85 to +0.15) Wm-2, while cloud-aerosol interactions are estimated at -0.45 (-1.2 to 0.0) Wm-2 (IPCC, 2013). The net impact is a cooling with an effective radiative forcing (ERF) of 0.9 (-1.9 to -0.1) Wm-2 (IPCC, 2013). One effect of aerosols which has not been well evaluated is their effect on atmospheric chemistry. Atmospheric aerosols provide a surface for homogeneous reactions to occur, altering reactions rates and the availability of oxidants, thereby influencing the removal/production of radiatively important species such as methane (CH4) and tropospheric ozone (O₃). Oxidants such as the hydroxyl radical (OH) determine the atmospheric lifetime and hence burden of CH4, therefore changes to atmospheric aerosols which impact oxidation chemistry will also influence RF due to CH4. This effect could enhance or offset the negative RF of aerosols, depending on how the individual aerosol changes availability of oxidants. Quantifying the importance of this mechanism for RF is necessary to provide accurate estimates of the effect of aerosols, and assess relative effectiveness of measures to decrease aerosol emissions and precursors.

Using a sophisticated aerosol micro-physics model (GLOMAP) coupled to the TOMCAT three-dimensional chemical transport model, we separately simulate changes in atmospheric composition resulting from a 50% decline in anthropogenic emissions of black carbon aerosol (BC), volatile organic compounds (VOCs) and anthropogenic precursors of sulphate and nitrate. The impact of changes to each aerosol on lifetime of CH4 is then calculated to establish the resulting impact on CH4 burden and RF. Cutting global anthropogenic SO_2 emissions by 50% decreases atmospheric sulpate concentrations by \sim 44% after 2 years, while increasing global OH concentrations by \sim 0.9%. CH4 lifetime is reduced by approximately 50 days as a result, leading to a decrease in CH4 burden of \sim 38ppb. NO_x is anticipated to have a similar but much larger effect (Matsui and Koike 2016). The Edwards and Slingo offline radiation model is also used to calculate changes to direct and indirect aerosol forcing. Presented here is the net RF change following 50% emission decrease of each aerosol or precursors, accounting for the direct and indirect aerosol effect as well as indirect effects via oxidation chemistry on the RF due to CH4 and tropospheric O_3 .