



## **Changes in the aerosol direct radiative forcing from 2001 to 2015: observational constraints and regional mechanisms**

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Accurate representations of anthropogenic emissions and their relationships to aerosol radiative effect are important to understand past and future climate. Over the 2001-2015 period, anthropogenic emissions have experienced large changes in both their speciation and geographic locations. Over this period, observations from CERES provide constraints on the outgoing shortwave clear-sky radiation ( $R_{\text{swcs}}$ ). From these observations, we estimate the change in shortwave clear-sky direct radiative effect from aerosols (SDRECS) by accounting for the impacts of ozone, albedo, and water vapor on  $R_{\text{swcs}}$  variability. We find that SDRECS increased (i.e., less radiation scattered to space by aerosols) over Europe ( $0.7\text{--}1\text{ W m}^{-2}\text{ dec}^{-1}$ ) and the US ( $0.9\text{--}1.8\text{ W m}^{-2}\text{ dec}^{-1}$ ) and decreased over India ( $-0.5\text{--}-1.9\text{ W m}^{-2}\text{ dec}^{-1}$ ). Comparisons with the GFDL chemistry climate model AM3, driven by CMIP6 historical emissions, show that changes over Europe and the US are well captured, which largely reflects the mature understanding of the sulfate budget in these regions. In contrast, the model overestimates the decrease of SDRECS over India and China. Over China, this bias can be partly attributed to the decline of  $\text{SO}_2$  emissions after 2007, which is not captured by the CMIP6 emissions. In both regions, we also find much larger contributions of nitrate and black carbon to changes in SDRECS than in the northern mid-latitudes, which highlights the need to better constrain their precursors and chemistry. Globally, our model shows that changes in the aerosol direct forcing between 2001 and 2015 ( $+0.03\text{ W m}^{-2}$ ) are dominated by black carbon ( $+0.12\text{ W m}^{-2}$ ) with significant offsets from nitrate and sulfate ( $-0.06\text{ W m}^{-2}$ ). Changes in the sulfate (+7%) and nitrate (+60%) direct forcing over the 2001-2015 period are poorly related to changes in their precursors (-12.5% and 19% for  $\text{SO}_2$  and  $\text{NH}_3$ , respectively), due to non linearities in their chemistry.