4.086 Reassessing the global secondary organic aerosol (SOA) budget and vertical distribution: stronger production, faster removal, shorter lifetime..

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Abstract:

Current chemistry climate models fail to reproduce the vertical distribution of organic aerosols (OA) with a large underprediction near the surface and a factor of 10-100 divergence in predicted OA profiles among models, reflecting our poor understanding of processes controlling the OA lifecycle. Recent laboratory and ambient measurements suggest that both production yields and removal rates of chemically produced secondary organic aerosols (SOA) are much stronger and more diverse than currently assumed in chemistry-climate models. In this study, we re-assess the global SOA distribution and budget with newly proposed SOA production and loss processes derived from these recent measurements, as well as from theoretical calculations. We evaluate and discuss the relative importance of increased production rates (wall corrected yields), chemistry of semi-volatile and intermediate volatility organic compounds, removal pathways for organic vapors and particles (e.g. dry and wet deposition, photo-dissociation, evaporation, and heterogeneous surface reactions), and their effect on the SOA vertical distribution and budget using a global chemistry-transport model. We compare simulated SOA from various model configurations against ground, aircraft and satellite measurements to assess the extent to which these new developments in our understanding of SOA formation and removal processes are consistent with observed characteristics of the SOA distribution. Our results show strong changes in predicted vertical profiles of organic aerosols with higher SOA concentrations in the boundary layer and lower concentrations in the upper troposphere, which appear to be in a better agreement with aircraft measurements.