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BROWNING: Brown carbon chemistry: modeling the uptake of ammonia on secondary organic aerosols and its effect on the radiative forcing

Description

Organic aerosols (OA) affect climate forcing and human health, but still large uncertainties exist in their sources and evolution. A large fraction of submicron particulate matter, 20%90%, can be attributed to OA. Recent studies have shown that secondary organic aerosols (SOA) account for a large fraction of the OA burden. Most OA efficiently scatter visible radiation and contribute to a cooling effect in the atmosphere. However, a significant fraction of OA absorbs radiation in the near-UV and visible ranges. This fraction is typically referred to as "brown carbon" (BrC). Like black carbon and mineral dust, BrC contributes to warming of the atmosphere. Recent studies have identified that some initially scattering SOA undergoes browning during exposure to reduced nitrogen compounds such as ammonia, ammonium from dissolved ammonium sulfate or other salts, and amino acids. The contribution of such light-absorbing OA to the total warming effect of BrC is still highly uncertain and may be significant in areas that have high emissions of ammonia or dissolved ammonium salts.

The main objective of this proposal is to explore the effects of the reactive uptake of ammonia by SOA on the optical properties and burden of anthropogenic and biogenic SOA using both experimental and modeling approaches. We propose implementing recently identified NH3SOA chemistry in a state-of-the-art atmospheric-chemistry model, the NMMB-MONARCHv1.0 model. The new system will allow us to quantify the fraction of BrC from primary and secondary sources, and the contribution of this light-absorbing SOA on the radiative forcing of OA. We will start with results of laboratory experiments at the University of California, Irvine and the U.S. Department of Energys Pacific Northwest National Laboratory on the chemistry and microphysics of NH3SOA chemistry. Global and regional modelling experiments will be conducted to explore the dynamics of secondary BrC formation and aging and advance the scientific communitys understanding of the role of BrC chemistry in air quality and radiative forcing. Complementarily, we will conduct an analysis of a recent field campaign conducted by CSIC in northeast Spain characterising the formation of secondary aerosols and their optical properties. An analysis of the measurements combined with the model results will provide valuable information on the presence of BrC and the relevance of the reactive uptake of NH3 on SOA in the real atmosphere.

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