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Using the Amazon as a natural laboratory to understand anthropogenic enhancement of biogenic SOA

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Understanding how anthropogenic emissions have modified natural biogenic secondary organic aerosol (SOA) formation constitutes one of the largest uncertainties in our understanding of the radiative forcing of climate. Due to ubiquitous influence of anthropogenic emissions over most terrestrial locations in the Northern Hemisphere, it is difficult to establish baseline biogenic SOA formation i.e. biogenic SOA that would be formed in the absence of anthropogenic perturbations. The vast Amazon rainforest during its wet season is one of the few remaining places on Earth where atmospheric chemistry transitions between preindustrial-like and present-day polluted conditions and serves as a unique natural laboratory to study anthropogenic-biogenic interactions. We develop insights from several laboratory measurements to simulate SOA formation in the wet-season Amazon using a high-resolution regional model (at 2 km grid spacing) and develop mechanistic insights about the role of anthropogenic emissions in biogenic SOA formation. We perform model simulations using the community regional Weather Research and Forecasting Model coupled to chemistry (WRF-Chem) at cloud-, chemistry-, and emissions-resolving scales i.e. at 2 km grid spacing. Sensitivity simulations that turn the urban emissions on/off are performed to quantify the impacts of anthropogenic-biogenic interactions on SOA formation. We evaluate WRF-Chem simulations using aircraft-based field measurements of SOA during the Green Ocean Amazon (GoAmazon 2014/5) field campaign. Our results show that urban emissions increase concentrations of nitrogen-oxides (NO_x), which cause increase in oxidant (ozone and OH radical) concentrations within the otherwise pristine Amazon. Increased oxidant concentrations catalyzed by NO_x substantially increase reactions of forest organic carbon, emitted as volatile organic compounds (VOCs include isoprene, monoterpene and sesquiterpene compound classes), and thereby enhance biogenic SOA formation by 60-200% on average in plume-influence regions. Model simulated enhancements agree with those observed by the aircraft which rapidly and concomitantly measures organic aerosols in background and plume-influenced locations using the Aerosol Mass Spectrometer (AMS). Our results provide a clear picture of how anthropogenic emissions might have substantially enhanced natural biogenic SOA formation since preindustrial times on Earth.