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Quantifying organic aerosol removal in the remote troposphere: Constraints on physical and chemical removal of OA provided by the ATom mission

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Organic aerosol (OA) is one of the major contributors to the PM2.5 burden both in the continental Northern Hemisphere and globally. Understanding its sources and aging is central to current air quality control strategies. For the remote troposphere, sparse in-situ data to date results in highly under constrained OA prediction models, with model diversity of up to three orders of magnitude in the recent AEROCOM-II comparison.

In the course of the recent NASA Atmospheric Tomography (ATom) set of aircraft missions, we have acquired four unique global datasets of submicron aerosol concentration and composition over the remote Atlantic and Pacific Oceans. In the remote FT OA and sulfate are the main components (about 0.3 μg sm-3 in total, fairly constant outside of continental outflow. However, OA in the remote FT exhibits a much higher average carbon oxidation state than in continental airmasses (OSc up to +1 compared to -1 over the continents), much higher than assumed in most models. This also suggests a fairly hygroscopic OA. Nevertheless, in the cleanest/most remote parts of the global free troposphere (FT), sulfate predominates. This is not captured by current global models and suggests an additional chemical removal of OA (and possibly continuing formation of sulfate).

Using several different hydrocarbon-ratio based photochemical clocks in combination with back trajectories to infer the age of the airmasses sampled during ATom, we estimate that the lifetime of OA in the remote UT (after most of the convective removal has happened) is of the order of 4

days. In contrast, for chemically inert black carbon, the estimated removal timescale using the same method is significantly longer (about a week), in general agreement with previous estimates of physical removal that are used in models. The significantly shorter OA lifetime suggests an additional, chemical removal mechanism. This provides a key constraint for modeling of OA in the FT, based solely on measurements. Both heterogeneous oxidation by OH and aerosol photolysis are possible pathways for OA removal that have been suggested previously. Sensitivity studies in CESM2 AND GEOS-Chem with updated chemistry and aerosol sources are used to constrain the relative importance of each pathway for OA removal during ATom.

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