

Overview of the MUCHACHAS Campaign: Preliminary Results

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The Multiple Chamber Aerosol Chemical Aging Experiments, or MUCHACHAS campaign, was a project jointly funded by EUROCHAMP and the U.S. EPA. The campaign focused on a simple hypothesis: *Oxidation by OH radicals of organic vapors associated with Secondary Organic Aerosols will significantly change the SOA loading and characteristics.* Specifically, we expect that lightly oxidized first-generation VOC products will yield secondary products with lower vapor pressures, thus increasing SOA levels, while more heavily oxidized later-generation products will tend to fragment into lighter OVOCs, thus reducing SOA levels. MUCHACHAS focused principally on the effect of OH on first-generation SOA produced from the reaction between alpha-pinene and ozone. Our objective was to exploit the complementary strengths of several chamber facilities in a unified scientific study. In this case AIDA, PSI, SAPHIR, and the Carnegie Mellon University chambers permit extensive examination of the temperature dependence, NO_x dependence, sensitivity to the photolysis spectral characteristics (fluence and intensity), and sensitivity to the presence or absence of an OH-radical scavenger during initial theSOA formation step. The key feature of the experimental design was to separate that initial SOA formation step from subsequent OH aging by turning on a continuous OH source only after the initial SOA formation was complete. The experiments are thus substantially different from “photo-oxidation” experiments with simultaneous exposure to ozone and OH radicals.

The combined experiments revealed a substantial influence by OH radicals: at near ambient SOA loading, aging by OH radicals increased the mass loading by a factor of between 2 and 4, with corresponding changes in the physical and chemical characteristics of the SOA. Specifically, mass changes were accompanied by a substantial drop in volatility, increase in hygroscopicity, enhancement of oxidized markers in aerosol mass spectrometer measurements, appearance of secondary oxidation markers, and loss of first-generation vapors. Changes were generally most pronounced at the highest temperatures and lowest aerosol loadings, consistent with predominant oxidation in the gas phase. The timescale for these enhancements was of order 1 hour at elevated OH, corresponding to of order 5-6 hours under typical ambient conditions; transformations continued for the duration of the experiments following this relatively sharp, initial burst. We therefore expect these findings to be relevant to chemical transport models at all spatial scales.