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Direct observations of within-canopy ozonolyis of sesquiterpenes in Amazonia

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Sesquiterpenes are a class of highly reactive hydrocarbons produced by vegetation that may serve many important biological functions such as acting as antioxidants and as semiochemicals. By reacting with oxidants like ozone and contributing to organic aerosols, the emission of sesquiterpenes from vegetation may have significant impacts on climate, but unfortunately, only a few ambient measurements of sesquiterpenes have been reported. While generally assumed to have extremely short atmospheric lifetimes of a few minutes due to oxidation by ozone, technical difficulties have so far prevented the detailed characterization of within-canopy sources and sinks. Consequently, little is known about the biological functions of sesquiterpenes within forests and the controls over ecosystem-scale sesquiterpene emission/oxidation dynamics. Here, we present the first direct observations of within-canopy sesquiterpene ozonolysis reactions obtained during the BrazilianAir 2010 field campaign in central Amazonia. Sesquiterpene concentrations at all heights were inversely related with ozone, suggesting that within-canopy ozonolysis reactions strongly influence the diurnal, vertical, and week to week variability in sesquiterpene concentrations. Unlike all other compounds investigated which showed strong diurnal patterns with maxima during midday, sesquiterpene concentrations peaked at night when ozone concentrations were the lowest. The low ozone atmosphere in the tropical rain forest biome at Biosphere 2 was used to demonstrate that these observations are unlikely to be explained by enhanced nocturnal emission rates; like other compounds observed, branch emissions and whole biome concentrations showed strong diurnal patterns with maxima at mid day, opposite of the pattern observed in the Amazon ambient air. In addition, unlike other primary compounds whose concentrations peaked within the Amazon canopy, sesquiterpene concentrations were highest near the ground, opposite to previously determined patterns for ozone. Finally, using ozone at 40m, the first attempt is presented to directly calculate within-canopy sesquiterpene ozonlysis rates. We conclude that within canopy sesquiterpene oxidation may be the dominant fate of biogenic sesquiterpenes emissions during the dry season (when ozone is high), thereby protecting plants from oxidative damage. Our findings should help to better understand the environmental and biological controls on the dynamics of chemical emission, loss, and transport of highly reactive species like sesquiterpenes within canopies and their potential biological and atmospheric impacts.