Bob Yokelson obtained his PhD in chemical physics at Yale University in 1991. From 1991-1993 he worked as a post-doctoral associate in atmospheric chemical kinetics at NOAA, focusing on the photochemistry and kinetics of stratospheric ozone depletion. Since 1993 he has been at the University of Montana in Missoula as an Associate Research Professor. His primary work has been global, ground-based, airborne, and laboratory measurements of reactive and stable constituents in emissions from biomass burning and vegetation and their downwind transformations. He has been involved in several projects to model the production and photochemistry of combustion and vegetative emissions, including work on cloud-processing. Recently his research group performed comprehensive emissions measurements for burning garbage and cooking fires in Mexico in spring 2007.

Airborne and ground-based measurements of fire and biogenic emissions during the 2004 Amazonian dry season

Robert J. Yokelson[1], Thomas Karl[2], Ted J. Christian[1], Paulo Artaxo[3], João A. Carvalho Jr.[4], Ernesto C. Alvarado[5], Jim Greenberg[2], Donald R. Blake[6] and Alex Guenther[2]

[1] Department of Chemistry, University of Montana, Missoula, Montana, USA
[2] Atmospheric Chemistry Division, National Center for Atmospheric Research, Boulder, Colorado, USA
[3] Department of Applied Physics, Institute of Physics, University of São Paulo, São Paulo, Brazil
[4] Department of Combustion Engineering, National Institute for Space Research, INPE, Cachoeira Paulista, Brazil
[5] College of Forest Resources, University of Washington, Seattle, Washington, USA
[6] Department of Chemistry, University of California, Irvine, California, USA

Volatile organic compounds (VOC) are key species in atmospheric chemistry because of their reactivity and roles as pollutants, precursors to ozone (O_3, an air toxic and greenhouse gas), and as greenhouse gases themselves. Fine carbonaceous particles also have important atmospheric impacts such as effects on radiative transfer (through light scattering and absorption) and cloud formation (by acting as condensation nuclei). The two largest sources of VOC and fine carbonaceous particles in the global atmosphere are biomass burning and vegetation [1]. Fires due to tropical deforestation account for much of the total global vegetation burning [2] and moreover, tropical forests produce a large part of the global biogenic emissions [3]. A substantial portion of the global deforestation is occurring in the Amazon basin driven by conversion to pastures, shifting cultivation, and intensive agriculture (e.g. plantations). Conversion is associated with both long-lasting changes in the vegetative emissions and large "pulses" of fire emissions. Intensive agriculture requires removal of all onsite woody material in relatively intense fires, while use for pasture and shifting cultivation can tolerate large amounts of residual woody debris (RWD). Therefore these latter uses are usually associated with a long-term series of smaller fires. The RWD subsequently contributes to emissions both through decomposition and by providing large amounts of fuel in post-conversion, site maintenance fires.

The Tropical Forest and Fire Emissions Experiment (TROFFEE) was affiliated with the Large Scale Biosphere-Atmosphere Experiment in Amazonia (LBA), an international cooperative research program led by Brazil, focused on producing new scientific knowledge on the functioning of Amazon ecosystems. TROFFEE was funded by the US National Science Foundation, and led by the Universities of Montana and São Paulo, the Brazilian National Institute for Space Research (Instituto Nacional de Pesquisas Espaciais, INPE), and the (US) National Center for Atmospheric Research (NCAR). The measurements in the Brazilian Amazon were made during the 2004 dry season (August-September). The field work was preceded by comprehensive laboratory measurements of the emissions from burning tropical fuels. During this phase we compared multispecies chemical analysis results from two instruments recently developed to minimize the common problem of chemical interference: an open-path Fourier transform infrared spectrometer (FT-IR) and gas chromatography (GC) coupled with a proton-transfer reaction mass spectrometer (PTR-MS). The results allowed us to determine "generic" branching ratios for compounds in smoke with the same mass-to-charge ratio. This means that the signal for various mass channels measured by the PTR-MS during the field campaign could be divided into the contributions from the individual VOC. One of the most important atmospheric VOC is formaldehyde (HCHO) and the PTR-MS response to HCHO is humidity dependent. The laboratory FTIR data for water and HCHO was compared to the PTR-MS HCHO and a humidity-dependent correction factor for the PTR-MS field HCHO data was determined. Finally, the lab work allowed us to probe some additional tropical fuels we did not sample in the field, for example sugar cane [4].

The TROFFEE smoke sampling [5]

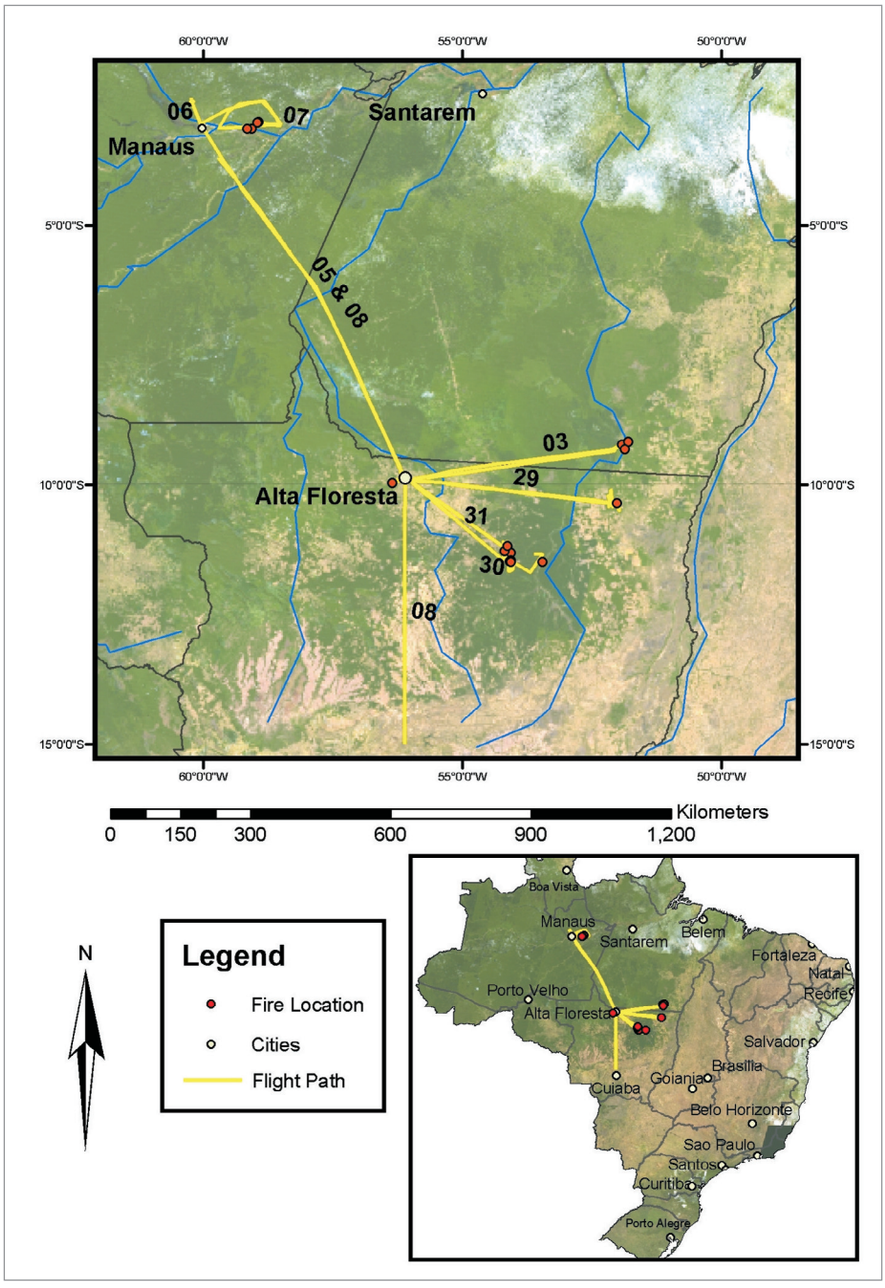

Figure 1. The Tropical Forest and Fire Emissions Experiment (TROFFEE): the flight tracks and the locations of the fires sampled in the Brazilian Amazon during the dry season of 2004 (August-September).

included the first-ever field measurements of the emissions of oxygenated VOC (OVOC) from tropical deforestation fires. The previously known VOC were hydrocarbons that react mainly with atmospheric oxidants such as the hydroxyl radical (OH). The OVOC are of great interest because they also photolyze (react with sunlight), can be precursors for OH, and are now known to be much more abundant in the atmosphere than previously believed. Our TROFFEE data showed that about 80% of the VOC emissions from tropical deforestation fires are OVOC, a much higher contribution than previously assumed for modeling purposes. Our particle (PM_{10}, particulate matter with an aerodynamic diameter of less than 10 μm) emission factor (average emission rate of given pollutant for a given souce) was 17.7 g kg^{-1}, which is about 25% higher than implied by previous reports for tropical forest fires and probably reflects both a trend towards, and the sampling of, larger fires than in previous studies. Comparison of fresh and aged smoke showed evidence of secondary gas phase production of O_3, organic acids, and acetone, but not of acetaldehyde and methanol. A significant fraction of the total burning in 2004 occurred within about 10 ideal lower humidity days and generated a mega-plume, greater than 500 km in width. We sampled one hour to one day old smoke in the mega-plume on its peak day (September 8). The mega-plume contained 10-50 ppbv of numerous reactive species such as O_3, ammonia, nitrogen dioxide, methanol, and organic acids and high PM_{10}. This is an intense, poorly understood, but globally important, chemical processing environment. The mega-plume was transported over São Paulo, Brazil (~1800 km from the source region) during September 14-19 and more than tripled the city's aerosol optical thickness on September 17.

Ground-based smoke measurements [6] with a new cart-based FTIR system showed that residual smoldering combustion of RWD (which produces initially unlofted emissions) can cause the estimated regional fire emissions of several reactive VOC to increase by 20–50% compared to estimations based only on airborne measurements in lofted plumes. Spot measurements of charcoal kiln emissions showed an increasing VOC/CO ratio in the emissions when the carbonization period was extended. We observed high emission ratios from burning dung for acetic acid to CO (~7%) and ammonia to CO (~9%). This has implications for secondary aerosol formation in regions such as South Asia where dung is an important biofuel. The INPE/UW team investigated the dynamics of fuel consumption and site recovery as well as fire effects on ground water and other ecosystem components.

In a relatively fire-free part of the Amazon basin, airborne and ground-based concentration measurements and flux measurements by eddy covariance and two different (mean and variance) mixed layer gradient techniques were used to assess the impact of isoprene and monoterpene emissions on atmospheric chemistry in the Amazon basin [7]. Average noon isoprene (7.3±2.3 mg m^{-2} h^{-1}) and monoterpenes fluxes (1.2±0.5 mg m^{-2} h^{-1}) from pristine forest compared well between ground and airborne measurements. The biogenic emission model MEGAN (Model of Emissions of Gases and Aerosols from Nature) predicts similar isoprene fluxes within the model uncertainty, but tended to underpredict the isoprene emissions from some plantations (e.g. soybean) that were sampled from the air. Isoprene

Figure 2. Instrumentation used in TROFFEE on the INPE Bandeirante aircraft: on the right PTR-MS (NCAR) and on the left Airborne FTIR and whole air sampling (University of Montana and University of California Irvine). In front O_3, PM_{10}, 3-wavelength nephelometry, GPS (University of São Paulo).

and monoterpenes accounted for ~75% of the total OH reactivity in this region suggesting that these species control the oxidative capacity of the tropical, and much of the global, atmosphere. The rate of photochemical oxidation of isoprene increased significantly within a ubiquitous, broken layer of fair weather cumulus. This may help explain why some global models tend to incorporate estimates of vegetative emission rates that are near the low end of measured values, yet still predict ambient concentrations of these species that are higher than many observations.

bob.yokelson@umontana.edu

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