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Emissions Relationships in Western Forest Fire Plumes: I. Reducing the Effect of Mixing Errors on Emission Factors

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. Abstract: Studies of emission factors from biomass burning using aircraft data complement the results of lab studies and extend them to conditions of immense hot conflagrations. We illustrate and discuss emission relationships for 422 individual samples from many forest-fire plumes in the Western US. The samples are from two NASA investigations: ARCTAS (Arctic Research of the Composition of the Troposphere from Aircraft and Satellites) and SEAC4RS (Studies of Emissions and Atmospheric Composition, Clouds, and Climate Coupling by Regional Surveys). This work provides sample-by-sample enhancement ratios (EnRs) for 23 gases and particulate properties. Many EnRs provide candidates for emission ratios (ERs, corresponding to the EnR at the source) when the origin and degree of transformation is understood and appropriate. From these, emission factors (EFs) can be estimated when the fuel dry mass consumed is known or can be estimated using the carbon mass budget approach. This analysis requires understanding the interplay of mixing of the plume with surrounding air. Some initial examples emphasize that measured $C_{\text{tot}} = \text{CO}_2 + \text{CO}$ in a fire plume does not necessarily describe the emissions of the total carbon liberated in the flames, C_{burn} . Rather, it represents $C_{\text{tot}} = C_{\text{burn}} + C_{\text{bkgd}}$, which includes possibly varying background concentrations for entrained air. Consequently, we present a simple theoretical description for plume entrainment for multiple tracers from flame to hundreds of kilometers downwind and illustrate some intrinsic linear behaviors. The analysis suggests a Mixed Effects Regression Emission Technique (MERET), which can eliminate occasional strong biases associated with the commonly used normalized excess mixing ratio (NEMR) method. MERET splits C_{tot} to reveal C_{burn} by exploiting the fact that C_{burn} and all tracers respond linearly to dilution, while each tracer has consistent EnR behavior (slope of tracer concentration with respect to C_{burn}). The two effects are separable. Two or three or preferably more emission indicators are required as a minimum; here we used ten. Limited variations in the EnRs for each tracer can be incorporated and the variations and co-variations analyzed. The percentage CO yield (or the modified combustion efficiency) plays some role. Other co-relationships involving nitrogen and organic classes are more prominent; these have strong relationships to the C_{burn} to O_3 emission relationship. In summary, MERET allows fine spatial resolution (EnRs for individual observations) and comparison of similar plumes distant in time and space. Alkene ratios provide us with an approximate photochemical timescale. This allows discrimination and definition, by fire situation, of ERs, allowing us to estimate emission factors.