4

Emissions Measurements from Vegetation Fires: A Comparative Evaluation of Methods and Results

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ABSTRACT

Fires in the open environment produce a diversity of combustion products. Special techniques are needed to characterize the emissions ranging from microcombustion-evolved gas analysis to airborne monitoring of the full-scale phenomenon. This chapter discusses the advantages and disadvantages of each, the use of data in models for full-scale fires, and provides a discussion of the measurement needs for more fully understanding the release of emissions on a global/temporal scale.

INTRODUCTION

Fires in biomass fuels emit a complex mixture of particulate matter and gases into the atmosphere. Globally, the diversity of combustion products results from wide ranges in fuel types and fire behavior induced by the large variations in ecological types and weather phenomena. Fires in tropical ecosystems consume as much as 80% of the total biomass burned (approximately 6×10^{15} g yr^{-1}) on a global basis (Hao et al. 1990; Crutzen and Andreae 1990). In contrast, fires in the U.S.A. have been studied in detail, but produce a small percentage of the total emissions (less than 2%-3% [Ward and Hao 1991]). Most estimates of global emissions have been based on a few observations from laboratory-scale test fires, ground and airborne measurements from tropical fires, or from detailed experiments conducted in different fuel types of North America. The purpose of this chapter is to discuss different sampling protocols and

scales of experiments used for characterizing emissions from biomass fires and the potential effect scaling may have on results.

For modeling atmospheric chemical reactions in the atmosphere, there is a need to know the temporal and spatial scales of release of emissions from biomass burning Even with advancements made in the use of satellite technology (Fishman 1991 Kaufman et al. 1992), measurement methods now in existence are not adequate to characterize the various sources. To develop emissions source strength on a regiona scale, dynamic models are needed for evaluating biomass consumption based or land-use patterns, burning practices, fuel models, weather history, numbers of fires and area burned.

COMBUSTION PROCESSES OF FIRES IN BIOMASS FUELS

The complex nature of free-burning fires has made both fire behavior and smoke emissions difficult to model from first principle considerations. Because of the complex interactions, it is important to have a clear understanding of the combustion process before attempting to model the fire/emissions phenomena. Fires in biomas fuels typically progress through three stages. First, a fire advances through a process of fuel preheating (the volatilization of free water and low boiling-point hydrocar bons), which is the beginning of pyrolysis. Second, a flaming combustion zone i formed in which the pyrolyzed products of hemicellulose, cellulose, lignin, and volatile hydrocarbons are rapidly oxidized. Third, a postflaming combustion mod (undoubtedly low oxygen content environment), called smoldering combustion, con tinues to produce aerosols and gases which do not enter the flaming combustion zone

The dynamics of the flame structure and the extent of smoldering combustion ar influenced by the distribution of different size classes of biomass particles and th packing density of the particles in the fuel complex. In addition, the moisture conter gradients within strata of biomass affect the reaction rate and ultimate temperatur within the oxidation zone. Technically, fires in biomass fuels range from lamina diffusion flames (candles) to highly turbulent flame structures (fast-moving, wind driven fires). Compounding this is the wind's influence on the transport of oxygen t the volatilized fuel gases.

An advancing flame front interacts with fuel and the atmosphere to produce smok which undergoes change in the atmosphere. The steps are as follows:

1. fuel preheating zone,
2. fuel vaporization zone,
3. variable fuel-to-oxygen ratio inside and outside the flame zone,
4. air entrainment or quenching zone,
5. combustion product equilibration zone in the free atmosphere, and
6. prompt processing of emissions occurs within zones of water condensatio (cloud processing).

These zones are often not clearly delineated. Some of the research we discuss was designed to answer questions concerning processes of emissions formation. Generally, sampling methods are designed to examine concentrations of emissions and ratios of emissions occurring outside the quenching zone in areas of gas temperatures <100°C. Airborne systems have been used to measure the emissions preceding and following cloud processing. Specialized experiments are needed to study the interactions between these six zones and the effect each has on the release of smoke emissions or a specific emission.

FUNDAMENTAL PARAMETERS AND DEFINITIONS

Rothermel (1972) and Albini (1976) describe a series of models useful for predicting characteristics of the flame front: flame length, flame depth, residence time, rate of spread, and rate of heat release. The source strength for a fire is closely coupled to the rate of spread and the rate of consumption of biomass by the fire. Generally, emissions released through the flaming consumption of biomass are more fully oxidized than emissions from smoldering combustion. The models of Rothermel (1972) and Albini (1976) do not describe the characteristics of the smoldering combustion of biomass fuels. Methods for estimating source strength of emissions are usually based on separate analyses of the flaming and smoldering combustion processes (Ward and Hardy 1991). Here, we will use a more generic model in evaluating important parameters for assessing source strength as follows:

$$q_{yn} = (EF_{yn})\, w_y r, \tag{4.1}$$

where q_{yn} = source strength of emission n, $g\,m^{-1}\,s^{-1}$;

n = specific emission, e.g., CO, CH_4, CH_3Cl, etc.;

EF_{yn} = ratio of the mass of emission n released to the mass of biomass consumed, $g\,kg^{-1}$;

w_y = mass of available fuel per unit area, $kg\,m^{-2}$;

r = rate of spread of the fire, $m\,s^{-1}$; and

y = if present, equals the phase of combustion, either flaming (f) or smoldering (s), if absent, the variable is assumed to apply for the fire.

Each of these variables needs to be evaluated to estimate the smoke emissions released from the burning of biomass. Often the resolution needed only requires a measurement of the total area burned by the fire; it is not necessary to quantify the rate of spread of the fire. In this case, when a general estimate of emissions released on a regional scale is adequate, EF_n, the biomass consumption per unit area, and the area burned by the fires are the only parameters evaluated. This technique is used in making estimates of the release of emissions to the atmosphere on a global basis. For purposes where a specific q_{yn} rate is needed, r must be quantified to describe the rate at which

the fire is moving into unburned fuel. Fire spread models of Rothermel (1972) and Albini (1976) can be used for this purpose or estimates of rate of ignition can be made from empirical observations.

The mass of available fuel per unit area, w, is equal to the total mass of biomass above ground times a ratio that is termed the *combustion factor*. The combustion factor is usually determined empirically or estimated from empirical data. Others have developed systems for evaluating the consumption of woody fuel, litter fuel, and duff (Brown et al. 1991).

The adoption of *carbon mass balance* (CMB) methods of ratioing the emissions released to the biomass consumed (Ward et al. 1979; Radke et al. 1990b) has expedited techniques for empirically evaluating EF_{yn}. Generally, laboratory methods (Nelson 1982) and some field techniques (Ward and Hardy 1991) use the method for quantifying EF_{yn} ratios for the flaming and smoldering combustion phases and for the carbon released during each combustion phase. The application of airborne systems for measuring average EF_n ratios has been greatly improved through the application of CMB methods (Radke et al. 1990b). There are only a few cases, however, where EF measurements have been made for the flaming phase using airborne systems (Radke et al. 1991; Einfeld et al. 1991). Usually, after the first few minutes of a fire, the emissions from the flaming and smoldering combustion processes become intermixed and it becomes difficult to assess which process (flaming or smoldering) is dominating the emissions being measured. An average EF_n ratio can be computed for the fire based on the carbon released during each phase of combustion:

$$EF_n = \frac{(EF_{fn})\, w_f + (EF_{sn})\, w_s}{w_f + w_s}.$$

(4.2)

Progress in modeling the release of emissions has come through the adoption of measurement of *combustion efficiency* (η) defined as the percent of carbon released in the form of CO_2 (Ward and Hardy 1991). In the combustion literature, η is based on the ratio of the actual heat released in a combustion process to the heat of combustion. The measurements of η based on either CO_2 or on heat released by the oxidation process are closely correlated. The η can be computed by:

$$\eta = \frac{[CO_2 a]}{[CO_2 t]} = \frac{CO_2 - c}{CO_2 - c + CO - c + HC - c + PM - c}$$

(4.3)

where $CO_2 a$ = actual concentration or mass of CO_2 released by the fire,
       $CO_2 t$ = theoretical limit of concentration or mass of CO_2 if all carbon is converted to CO_2,
       $CO_2 - c$ = mass of carbon or molar concentration of carbon where "$-c$" denotes carbon content of the molecular species for CO_2, CO, hydrocarbon (HC), and particulate matter (PM).

A convention in practice is to correlate dependent variables with the independent variable CO/CO_2. This ratio is correlated with η ($r = 0.99$) over a narrow range of η from 0.8 to 1.0. The CO/CO_2 ratio can be normalized by dividing the CO_2-c by the sum of carbon contained with the CO and CO_2; it becomes linearly correlated with other products of incomplete combustion as proposed by Ward and Hao (1991). They proposed that this parameter be defined as the modified combustion efficiency ($\hat{\eta}$) as follows:

$$\hat{\eta} = \frac{CO_2 - c}{CO_2 - c + CO - c} .$$

(4.4)

Figure 4.1 shows the CO/CO_2 ratio, η, and $\hat{\eta}$, correlated with CO_2 concentration. Hence, the more fundamental variable is recommended for more general use in correlating carbon emissions to a property of the combustion system. The $\hat{\eta}$ has been used as an estimator of η from the data of Ward and Hao (1991) where,
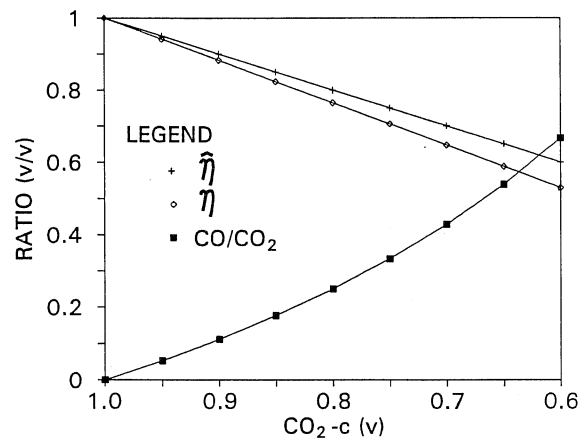
$$\eta = \frac{(\hat{\eta} - 0.15)}{0.85} .$$

(4.5)

Figure 4.1 A comparison of the linearity of the ratio of CO/CO_2 to CO_2 concentration., combustion efficiency (η), and modified combustion efficiency ($\hat{\eta} = CO_2$-c/[CO-c+CO_2-c]).

METHODS USED TO MEASURE EMISSIONS RELEASED FROM VEGETATION FIRES

Extremes in scale of measurement are often used to circumvent the hostile environment of the open burning fire and to reduce the variance encountered in the range of natural conditions. Methods being employed range from satellite optical-depth and numbers of fires measurements to very small controlled combustion experiments.

Laboratory and microcombustion techniques have been used on one end of the scale, airborne and satellite technology on the other. The next section discusses the smallest scale and moves progressively to the largest-scale modes of measurement. Table 4.1 summarizes examples of research performed in the laboratory using milligram- to kilogram-sized samples of biomass fuel and makes comparisons to systems used in the field to characterize the full-scale to near full-scale phenomena.

Table 4.1 Scale of fire studies in developing and characterizing emission from fires in biomass fuels.

Advantages	Disadvantages
Microcombustion Systems	
Pyrolysis products	Scale
Mechanisms	Fuel arrangement
Replication of experiments	Reaction pathways
Environment	Quenching of reactions
Temperature of fuel	Vessel walls
Flow-Reactor System	
Scale	Reaction pathway
Closed system	Flame scale
Controlled environment	Wall losses
Quenching	Fuel arrangement
Diffusion and/or premixed flames	
Combustion Laboratory (kg quantities of fuel)	
Scaling similarity for low intensity	Wall losses
Modeled fuel beds	Reaction pathway
Lifted fuel beds	Sample zone temperature
Lighting similarity	Costly to maintain
Replicated studies	Fuel moisture gradients
Environment control	Representativeness
Quenching control	
Complex train of analyzers (gc/ms)	
Field (Surface-based) (Small area and piled material studies)	
Near full-scale experiment	Temperature limitation
Replication of tests	Height limitation
Less costly than aircraft	Reaction pathway
Fuels, fire behavior, CMB	
Field (Airborne) (Full-scale fires and large areas)	
Full-scale	Weather (clouds)
Plume dynamics (trajectory analysis)	Lower limit of plume
Multiple sensors in same sample space	Mountainous terrain
CMB	Large test areas
Cloud/emissions interaction studies,	Costly
	Replication is difficult
	Fuel and fire behavior documentation

Microcombustion

In one microcombustion experiment, Clements and McMahon (1984) processed 10-mg samples of ground-up pine needles to evaluate emission factors for total hydrocarbons (THC), CO, CO_2, volatile organic compounds, and thermoparticulates. The matrix of tests included oxygen and non-oxygen (nitrogen) environments and simulated pyrolytic processes associated with smoldering combustion. They found that EF_{PM} values averaged 85 g kg^{-1} in an oxygen environment ($\eta = 0.67$). In another experiment, using the same apparatus and techniques of thermogravimetric-evolved gas analysis, Clements and McMahon (1980) found the production of oxides of nitrogen (NO_x) to be proportional to the level of fuel-nitrogen content and to agree well with values (NO_x emission factors range 1 to 10 g kg^{-1}) found from larger-scale fires. They concluded that most of the NO generated resulted from prompt release of bound nitrogen from the fuel substrate.

Temperatures in excess of 1200-1400°C are needed to disassociate significant amounts of molecular nitrogen to form NO. The potential for NO generation from dry, upland savanna types burning under high ambient temperature, low relative humidity, and brisk wind conditions needs to be investigated. Other high-intensity wildland fires may result in higher-than-normal combustion temperatures and the disassociation of molecular N_2 in larger quantities than would normally occur for lower-intensity fires.

Kuhlbusch et al. (1991) studied the nitrogen balance and found that only a small percentage of total fuel nitrogen could be accounted for by measuring NO_y. By using an argon or helium atmosphere, they deduced that an average of 36% of the fuel nitrogen is released in the form of molecular nitrogen, and most of this is released during the flaming phase of combustion.

Benner et al. (1977) used a closed combustion vessel for testing the photochemical potential of smoke from the burning of biomass fuels. They found that non-ozone oxidant formation always preceded ozone formation when smoke from small samples of pine needles was irradiated with ultraviolet light.

Edye and Richards (1991), using an inverted funnel and radiant heated petri dish, studied smoke emissions from smoldering needle and broad-leaved fuels for the emissions of phenol and furan compounds. Using CMB methods, the research is currently comparing early preliminary findings to larger-scaled systems of combustion.

Ward (1979) theorized that volatile materials contained in the forest fuel complex contribute significantly to the emissions produced, and that oxygen content of the fuel is inversely correlated with PM emissions. A flow reactor-combustion chamber system was developed for burning single and multiple component fuels introduced into the combustion chamber in a gaseous phase. Particulate matter emission factors for fuels of methane, propane, heptane, benzene, ethanol, ethylene glycol, and α-pinene were evaluated. A model was advanced suggesting that PM emissions for laminar-diffusion flames are a function of the flame envelope volume. Wildland fires

are a turbulent diffusion flame process. A system is now under development to pyrolyze solid fuels into a controlled environment combustion chamber, so that emissions tests can be performed for fuels of different chemical composition under controlled conditions (Richard Corlett, University of Washington, pers. comm.).

Highly controlled combustion experiments can be very effective in confirming hypotheses concerning processes affecting the release of emissions. In addition, these systems may be useful in establishing response functions for the release of various emissions as a function of combustion efficiency or other variables that then can be adjusted to the full-scale phenomena through a limited field research project.

Controlled Environment Combustion Laboratory

Feldstein et al. (1963) and Darley et al. (1966) pioneered concepts needed for evaluating emissions from combustion experiments using closed funnel devices for channeling and concentrating emissions. Fritschen et al. (1970) and Vines et al. (1971) used micro combustion hoods for burning small samples of forest fuels and deduced the approximate mixtures of carbon containing gases and PM. These data were used to optimize airborne sampling systems. Lobert et al. (1991) and Jenkins et al. (1991) used variations of the open-hood system for studying emissions from fires in different species of grasses and other fuels. Controlled environment combustion laboratory experiments have been perfected whereby kilogram-sized samples of forest fuels are arranged (including different levels of loading, packing ratios, moisture content, etc.) in fuel baskets on weighing platforms and burned; the emissions are exhausted through a hood-stack arrangement. McMahon and Tsoukalas (1978) studied the emissions of PM and polynuclear aromatic HC from the combustion of pine needle fuels burned using simulated heading fires (where the fire ran upslope) and backing fires (where the fire backed down slope). They found very high benzo[a]pyrene to PM ratios for backing fires (98 to 274 μg g^{-1} PM) with much lower values for heading fires (2 to 3 μg g^{-1} PM). These results were not substantiated under field research conditions and no explanation for the discrepancy was reported (White 1987; Ward 1989).

Ward et al. (1980) concluded that similarity of scaling of the fuel bed and flame structure of the combustion laboratory fires to that of the field was necessary to achieve agreement between results. More recent results of Ward and Hao (1991) comparing controlled environment combustion laboratory and combustion hood experiments suggest that η is the principal controlling variable and ratios of products of incomplete combustion remain constant over a wide range of η conditions (Ward et al. 1992; Weiss et al. 1991).

Griffith et al. (1991) used a Fourier transform infrared (FTIR) system in a combustion laboratory and in field studies to compare emissions of compounds that are difficult to sample and quantify, e.g., NH_3, N_2O, and CH_2O. The results were consistent between the field and laboratory for low-intensity smoldering combustion fires.

Sandberg et al. (1975) developed an inverse relationship between PM emissions and the rate of heat release by fires in modeled biomass fuel beds. They studied both the emissions of PM and HC as a function of treatment with a flame retarding chemical, diammonium phosphate (DAP). This study showed no significant reduction in PM emissions as a result of treating with the chemical. On the other hand, Philpott et al. (1972) found a significant increase in PM emissions using DAP, but a decrease in the total PM emissions resulting from the treatment of biomass with ammonium sulfate flame retarding chemical.

Opportunities exist for designing experiments that can be used for establishing response functions for emissions that are difficult to characterize under field conditions. We have used combustion laboratory methods to study the effect of chloride salts on the release of CH_3Cl. Reinhardt and Ward (unpublished manuscript on file at IFSL, Missoula, MT) studied emissions of CH_3Cl using a combustion hood device and concluded that most CH_3Cl is released during the smoldering combustion of biomass fuels and is correlated to the content of the Cl ion in or on the biomass fuel particles.

Advantages of using combustion laboratory facilities include the opportunity to study emissions released as a function of flaming and smoldering combustion using sophisticated instrumentation. In addition, the components of a fuel complex (fine fuels, coarse fuels, leaves, twigs, etc.) can be stratified and mixed to model natural fuel complexes. A disadvantage is the inability to duplicate the full-scale phenomena except for the lowest intensity fires.

Ground-Based Sampling

Sampling near the fire at downwind locations (Fritschen et al. 1970; Bonsang et al. 1991) or sticking a sample probe out over a fire (Crutzen et al. 1985; White 1987; Bonsang et al. 1991) will not always provide representative samples of the flaming and smoldering phases. Sampling must account for the rapid rate of fuel consumption during the flaming phase (Ward et al. 1992) and weight the emissions produced during the combustion phase by the carbon released in each phase. Ideally, to evaluate an emission factor or ratio that is representative of both the flaming and smoldering combustion phases, the emissions must be sampled at a rate proportional to the rate of carbon release over the duration of the fire. The emissions are usually sampled by phase of combustion.

Ward and Hardy (1991), using towers and cable systems to support sampling apparatus over fires, sampled the rate of release of carbon and emissions for a wide range of fuel and weather conditions. These data illustrate the dependency of carbon-based combustion products on η (Fig. 4.2). Ward et al. (1992) are using portable, computer controlled, Fire Atmosphere Sampling System (FASS) packages containing real-time analyzers for sampling CO_2, CO, NO, 3-dimensional winds, temperatures near the ground and near the sample inlets, and other variables at an adjustable rate of sampling from once per second. The systems are positioned in the fire with the
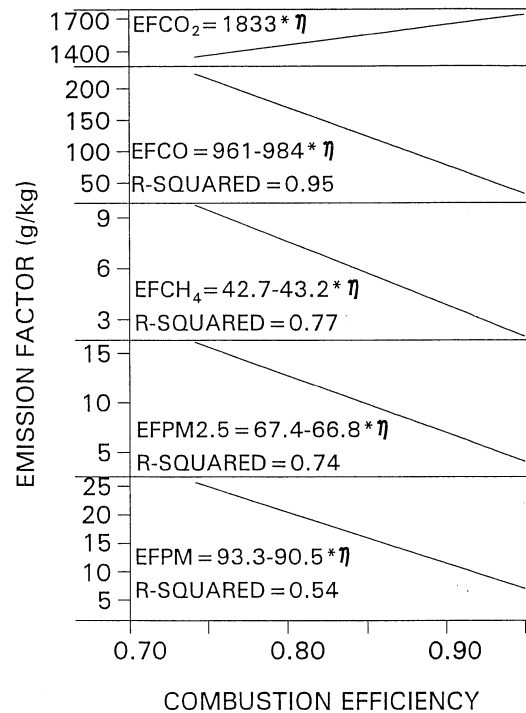
Figure 4.2 Relation of emission factors for CO_2, CO, and CH_4 particles less than 2.5 μm diameter and total particles without regard to size with combustion efficiency (Ward and Hardy 1991). Data are for prescribed fires in the states of Washington, Oregon, and California, U.S.A.

primary sampling head positioned 3–10 m above the flames. The FASS packages are triggered automatically by an increase in temperature or an increase in CO above a threshold value. The in-fire location avoids the potential for edge effects. Figure 4.3 shows models for emissions of particles less than 2.5 μm diameter (PM 2.5), CO, CH_4, and H_2 for fires in tropical ecosystems. The models compare favorably with those of Ward and Hardy (1991) for logging slash fires of the Pacific Northwest (U.S.A.); see Fig. 4.2.

White (1987) studied emissions of CO, PM, and B[a]P from the combustion of biomass fuels in the southeastern U.S.A. His hand-held apparatus was used near the flamefronts for sampling emissions of particles onto filters and gas into bags for spreading fires—both heading and backing fires. The measurements of B[a]P emissions (ranging from 7 to 58 μg g^{-1} of PM) were considered more accurate than the laboratory experimental fires of McMahon and Tsoukalas (1978). Ward (1989) corroborated this, reporting emission ratios for prescribed fires in logging slash of 13 ± 7 μg B[a]P per g of PM.
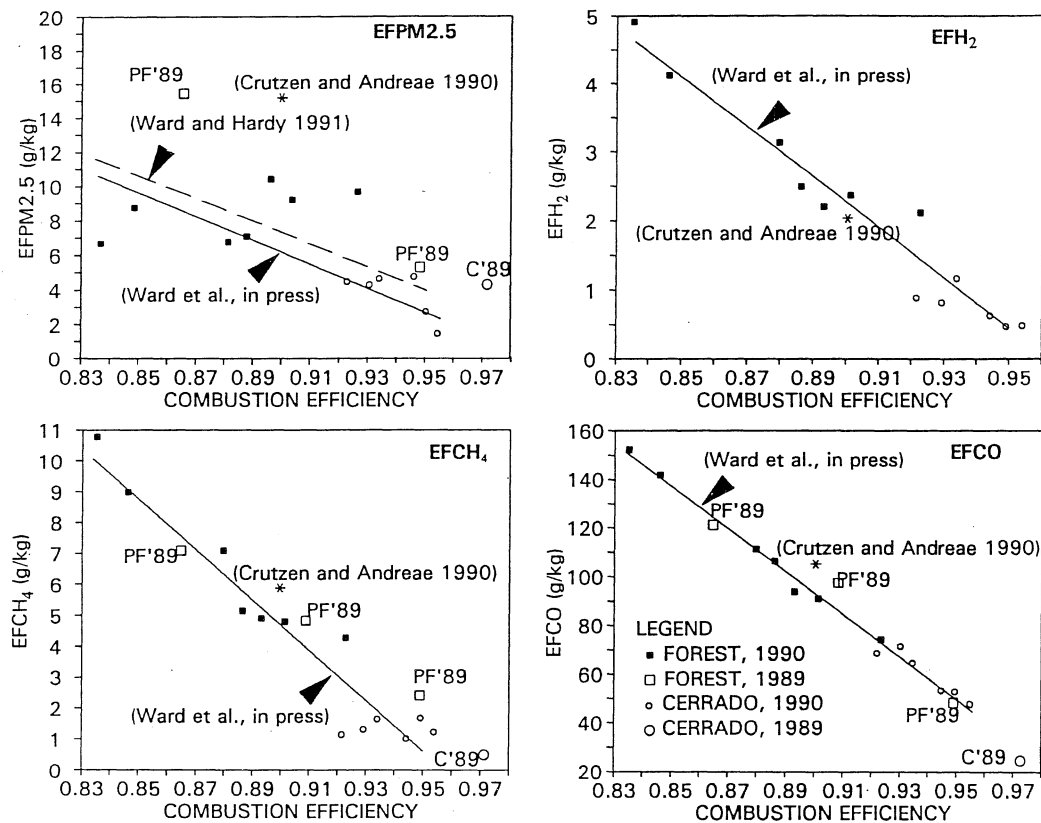
Figure 4.3 Models developed for combined forest and grassland fuels of Brazil (Ward et al. 1992). A comparison is made with average emission factors computed from Crutzen and Andreae (1990) and models for fine particles proposed by Ward and Hardy (1991).

A few experiments have been performed where surface sampling from towers has been coordinated with samples collected using airborne sampling techniques (Ward et al. 1979; Ward and Hardy 1991; Radke et al. 1990b). In general, results from both techniques compare favorably. Both tower and airborne sampling systems have profited from the use of CMB methods for characterizing the fuel consumed while producing the measured emissions (Ward et al. 1979; Radke et al. 1990b).

Field sampling has many advantages including the opportunity to sample emissions from natural fuel complexes burned using full-scale burning techniques and the opportunity to characterize the fuels and fire behavior associated with the measured emissions. The experiments can be difficult to set up, relatively expensive, and for very high-intensity fires, the samples may be affected by the high temperatures.

Airborne Sampling

Airborne sampling of emissions from vegetation fires has proven extremely useful primarily because of the ability to transport many different instruments to fires of known characteristics and because of the range of scale of fires that can be examined. Sampling of the full-scale phenomena is also necessary for verification and interpretation of findings from research using scaled-down models of fires. Airborne and/or remote sensing sampling techniques are the only methods that allow for measurements of smoke emissions from high-intensity, fast-moving wildfires. Measurements of emissions from a large range of fires can be collected quickly with minimal intrusion (Crutzen et al. 1979; Stith et al. 1981; Crutzen et al. 1985; Radke et al. 1988; Cofer et al. 1988; Andreae et al. 1988; Vines et al. 1971; Radke et al. 1991; Ward et al. 1979; Westberg et al. 1981; Fritschen et al. 1970).

Vines et al. (1971) computed emission ratios of PM to CO_2 and performed flux measurements from several fires of known rates of fuel consumption to estimate EF_{PM} ratios. The CMB methods for field emission factor measurements were tested for ground-based and airborne methods of estimating emission factors using several methods, including CMB (Ward et al. 1979). The use of CMB methods for airborne sampling was perfected by Radke et al. (1990b) in the early 1980s. Emission factors and ratios of emissions have been important in studying atmospheric chemistry phenomena associated with emissions from biomass fires. Generally, however, the relation of emission measurements made from airborne systems has required extensive surface measurements of fuels and emissions for interpreting the fire behavior/fuel chemistry effects on emissions production (Ward et al. 1992).

Airborne measurements of ratios of species of HC were made by Westberg et al. (1981) to demonstrate the change in the composition of reactive HC with an increase in the concentration of O_3. They were able to show the rapid change of NO to NO_2 and the preferential loss of the more photochemically reactive HC. Others have measured the increase in O_3 in plumes from forest fires (Kaufman et al. 1992; Radke et al. 1991; Stith et al. 1981; Evans et al. 1977).
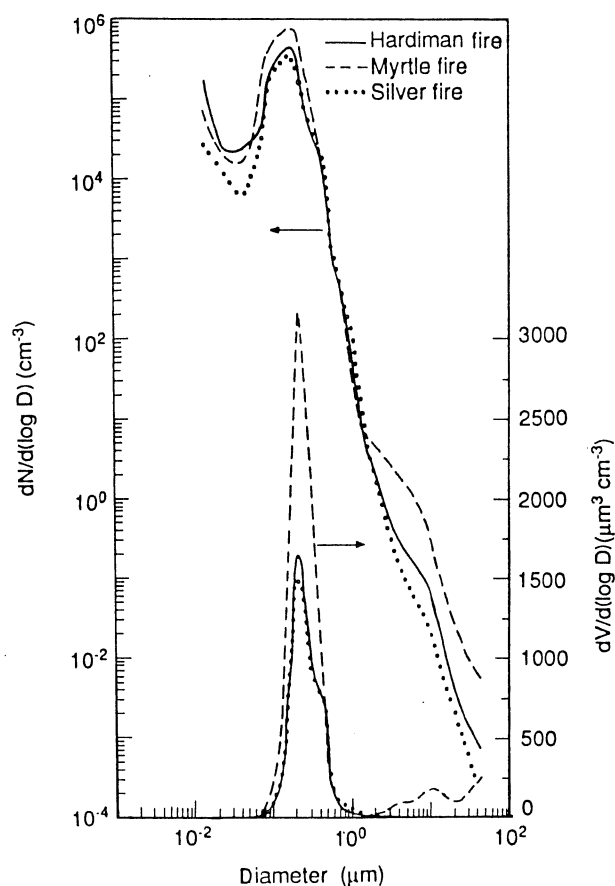
Figure 4.4 Average number and volume distributions of particles by size class for smoke plumes of a prescribed fire in Ontario, Canada (Hardiman Fire) and two wildfires in Oregon, U.S.A. (from Radke et al. 1991).

The physical properties of aerosols have been studied by Radke et al. (1991) for a wide range of fire conditions. Figure 4.4 illustrates the number of particles by size class over an extended range of diameter from 0.02 to 48 mm. The Myrtle Fire was a very active wildfire with a strong convection column during the time of sample collection; the Silver Fire was a very large fire, but was relatively quiescent during the periods of sample collection. For the Silver Fire of southern Oregon, U.S.A., Radke et al. (1991) performed a Lagrangian study to examine the downwind change in the particle size distribution over a 2-day period. The results showed an increase in particle size and a decrease in the number of particles as the plume moved downwind. The rate of change of the particles was somewhat faster than simple coagulation modeling would suggest. Perhaps, the strong positively charged cloud

observed over biomass fires may be an important factor in determining the rate of coagulation of particles (Latham 1991).

Improved methods are available to measure the flux of emissions and to validate models for predicting the flux of emissions from biomass fires. Equation 4.1 is a technique for estimating the flux of an emission (source strength, q). The q for PM and other emissions can be measured directly if airborne systems can measure the concentration within a cross-section and if the wind speed profile can be measured. Such direct *in situ* measurements have been made using multiple passes of an aircraft through the plume from top to bottom (an example is shown in Fig. 4.5; Stith et al. [1981] provide a detailed example).

An approach which greatly increases the efficiency of measurement utilizes airborne lidar. Here the entire plume cross-section (PM) can be measured with a single pass of the aircraft above the plume or on smaller scales, a single scan from a ground-based lidar (Uthe et al. 1982; Radke et al. 1990a; Brock et al. 1990; Banta et al. 1992; and Radke et al. 1992). Figure 4.6 shows a cross-sectional concentration drawing of an elevated wildland fire smoke plume (Radke et al. 1990a). Work now completed relates the intensity of backscatter measured with an airborne lidar system to the mass concentration of the aerosols (A. Waggoner, University of Washington, pers. comm.). With available lidars and dropwindsonde equipment, a long-range aircraft should be capable of measuring the net PM flux on continental scales (ignoring losses due to cloud processing, etc.). Such a capability is a vital part of verification of other predictive methods to estimate the flux of emissions from very large, distributed set of biomass fires on continental scales.
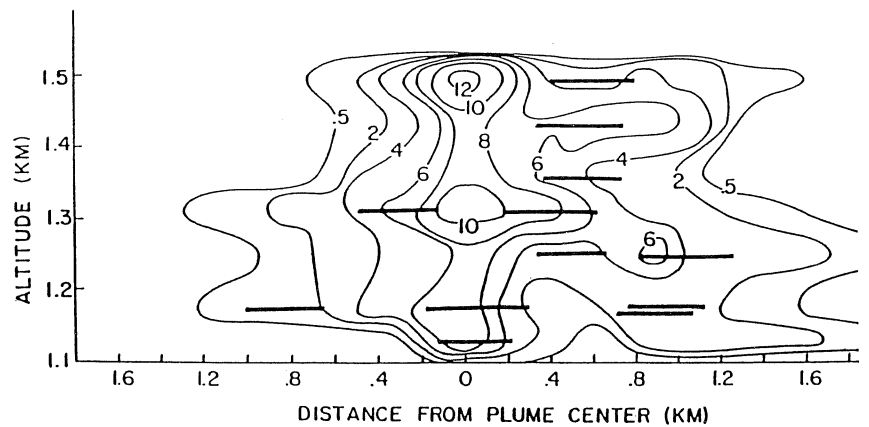

Figure 4.5 Smoke plume cross-section of the light scattering coefficient ($x \cdot 10^{-3} \cdot m^{-1}$ at 550 nm) measured perpendicular to the long axis of the plume using an integrating nephelometer. Airborne sampling was performed at multiple altitudes, and regions of equal values of light scattering coefficients were drawn from the measurements.

Figure 4.6 Example of LIDAR sounding of the backscatter from smoke particles in a plume from a prescribed fire in southern British Columbia, Canada. New relations provide for the drawing of isopleth lines of equal particulate matter concentration from data collected during a single pass either above or below plumes from biomass fires.

Radke et al. (1991) have conducted unique studies examining the effect of water cloud processing and scavenging on the physical and chemical characteristics of smoke particles in plumes from fires in biomass fuels. These processes are particularly important in assessing the atmospheric impact of large fires since a significant fraction of large fires have plumes capped by cumulus clouds, which frequently precipitate. Thus, a major fraction of the wildland fire smoke is exposed to cloud processing. To evaluate processes occurring within the condensing cloud, several techniques are used, including:

1. measuring the characteristics of the plume going into the cloud, then measuring the same features as the emissions exit the cloud;
2. measurements of the effect of cloud processing on emission factors measured below the cloud and on exit from the cloud;
3. measurement of the ratio of smoke or a surrogate for smoke interstitial to the cloud droplets; and
4. measurement in close time proximity of the smoke concentration going into the cloud and the amount of smoke in the cloud water.

Most work performed by Radke et al. (1991) have used method 1. Radke and Ward (1991) discuss the implications of emissions modification from cumulus cloud processing and how prescribed fire may be modified to enhance the cloud processing benefits from an air quality standpoint. Their work shows that precipitating clouds remove nearly 100% of the supermicron smoke particles in all, but the smallest capping cumulus clouds. In addition, the accumulation mode smoke particles (which represent the bulk of the smoke particles with potentially long atmospheric residence times) can enter cloud water with significant efficiency (40%-80%) and are removed from the cloud with equal efficiency (30%-90%) by precipitating cumulus with depths greater than 2 km (Figs. 4.7 and 4.8).

Airborne sampling techniques are expensive, but large volumes of data can be generated that reflect the effect of atmospheric processing on the released emissions. Measurements from airborne sampling platforms should be directly correlated with
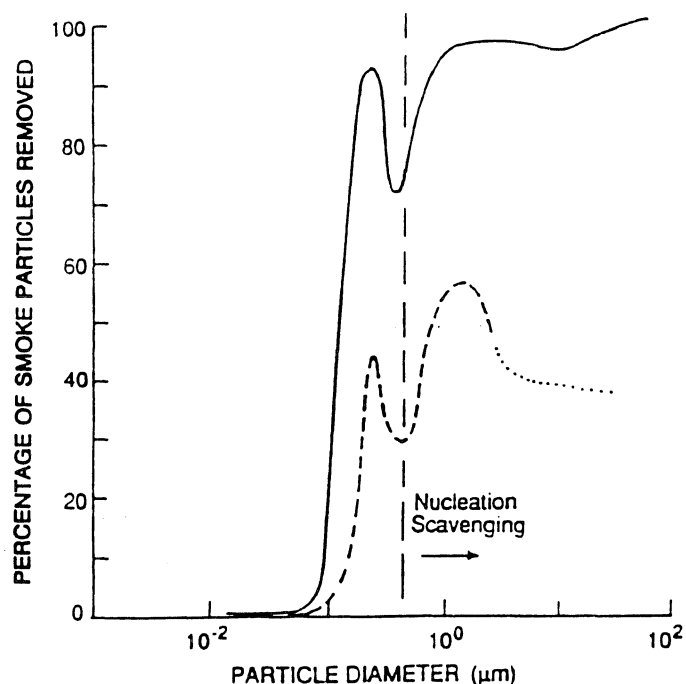
Figure 4.7 Two measurements of the percentage of mass of smoke removed by cloud and precipitation scavenging as a function of smoke particle size for the Battersby Prescribed Fire of Ontario Province, Canada (Radke et al. 1991).

spaceborne measurements. The capability of integrating samples across large areas makes airborne sampling especially attractive.

Satellite Techniques

There is a need to develop better methods of measuring the spatial and temporal scale of biomass burning. Several parameters need to be quantified to make satellite techniques a viable method for estimating emissions production. For example, given that total biomass per unit of area data are available continentally or regionally, some measurement of the combustion factor must be developed. In North America, a few techniques for estimating the combustion factor have been used; they depend on quantifying the amount and duration of precipitation and drying conditions (Brown et al. 1991). As with other techniques for estimating emission factors, knowledge is needed regarding η for the conditions of the ecosystem and the meteorological conditions affecting the fire. A variety of remote sensing techniques are being explored.

Kaufman et al. (1992) discuss the use of satellite technology for assessing fires and the smoke from fires. Fires can be detected using the Advanced Very High Resolution
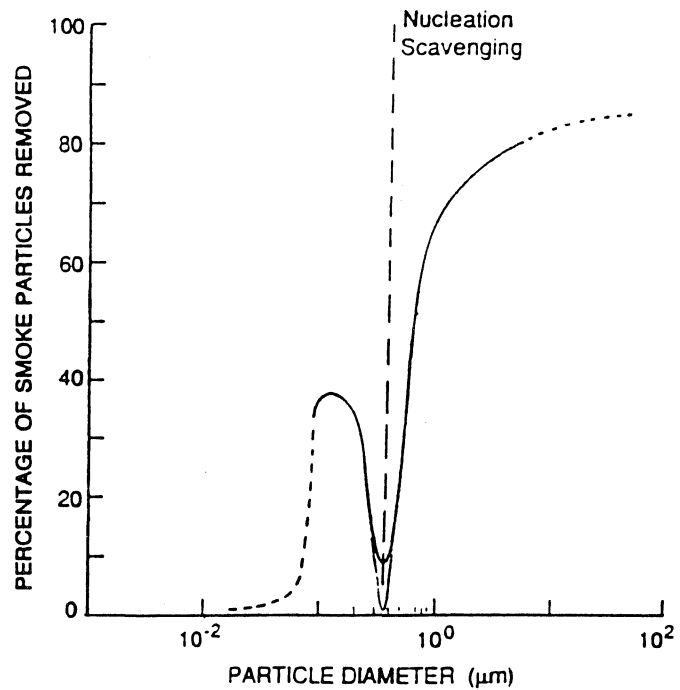
Figure 4.8 A measurement of the percentage of mass of smoke removed by cloud and precipitation scavenging as a function of smoke particle size for the Hardiman Prescribed Fire of Ontario Province, Canada (Radke et al. 1991).

Radiometer (AVHRR). This technique was developed at the Brazilian institute for space research (INPE). The 1 km resolution imagery for the 3.75 μm and 11 μm channels can sense fires as small as 10 m by 10 m (although it provides no measure of fire size at scales below 1 km^2). The AVHRR imagery is being used to monitor the frequency of burning in Brazil. For example, in 1990 in Brazil, there were 283,800 fires detected using this system from June 1 to October 1. In 1991, during the same period, there were 440,080 fires detected, or 55% more fires in 1991 than in 1990 (M. Luduvice and C.M. Mello Rosa of IBAMA, pers. comm.). A correlation has been established between the number of fires and the amount of deforestation as well as the fire count and the mass of emitted smoke particles (Kaufman et al. 1992). A combination of the Landsat Multi Spectral Spectrometer (MSS) satellite data has been used through a double sampling procedure to account for the area of deforestation through burn scar analysis and fire counts.

Kaufman et al. (1992) developed a technique for approximating the particle mass loading of the atmosphere from optical thickness measurements from AVHRR visible and near IR images taken over vegetated terrain. The correlation of the smoke particles with trace gases then makes it possible to estimate the release of trace gases

to the atmosphere from the biomass fires. In the future, correlations may be possible between estimates of O_3 concentrations and the quantity of emissions produced from biomass burning (Fishman 1991) and through the use of higher resolution multispectral imaging which may significantly improve estimates of both optical depth and PM mass loadings.

Spaceborne measurements are likely the only viable alternative for tracking the temporal distribution of the frequency of fires in many areas of the world. Coupling spaceborne, airborne, towers, laboratory, and benchtop studies provides a wide matrix of tools for examining fires from the process-level to the global-level. Interpreting results from each scale of measurement is providing the fundamental knowledge needed to examine the impact of smoke emissions on a global-to-continental and subcontinental scale.

INTERCOMPARISON OF PARTICULATE MATTER DATA

We have discussed many of the major advantages and disadvantages of characterizing emissions from wildland fires using various sampling techniques. The η has been suggested as a link between different scales of combustion systems to adjust the data from research-scale to full-scale phenomena. Although this seems to work in some cases (Ward and Hao 1991), it is probably only one factor affecting the release of many emissions. To demonstrate some of the problems, we have summarized PM emission factors for total PM (no size segregation) and for fine particles (generally less than 5 μm diameter) in Figs. 4.9 and 4.10 as a function of η.

For fine particles, the unpublished data of Reinhardt and Ward (master's thesis, Univ. of Washington) suggests a different response function with a slope coefficient 5 times larger than the data of Ward and Hardy (1991) and Ward et al. (1992). The data of Radke et al. (1991) using airborne sampling techniques is different as well; however, the data of Einfeld et al. (1991) fit the data of Ward and Hardy (1991). These differences, in part, are due to different fuel types and techniques of burning.

The samples of Reinhardt and Ward were collected using a combustion hood for the purpose of studying processes affecting the release of CH_3Cl, including the Cl content of forest fuels and processes of flaming and smoldering combustion. The fuel beds were spiked with NaCl and sprayed with water vapor to enhance smoldering combustion over flaming, which may partially explain the steep slope of the response function (Fig. 4.9).

On the other hand, the data of Ward and Hardy (1991) were collected within 5–15 m of the tips of the flame structure where the temperature occasionally exceeded 100°C. In contrast, the results of Radke et al. (1991) and Einfeld et al. (1991) were developed from airborne samples of emissions from wildland fires. The results from Ward and Hardy (1991), Radke et al. (1991), and Einfeld et al. (1991) are dependent on assessing the differential background concentrations of CO_2 and other gases for the CMB to provide accurate results. The in-plume-entrained air, from near the
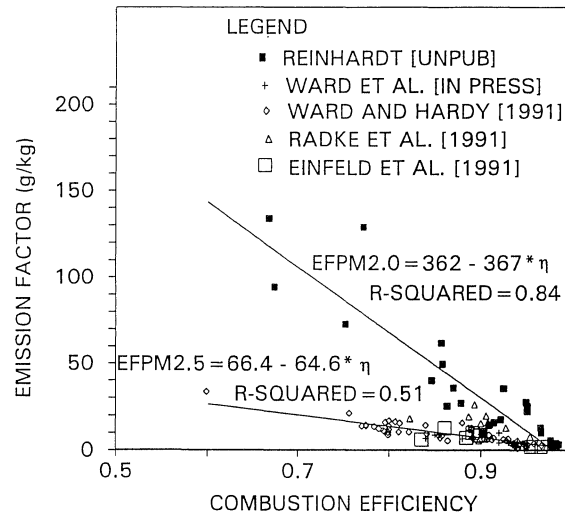
Figure 4.9 Models for predicting emission factors for fine particles (mass of particles less than 2.5 mm diameter) for a range of fire conditions with combustion efficiencies ranging from 0.6 to near 1.0. Data are intercompared with those of Radke et al. (1991) and the unpublished data of Reinhardt and Ward (on file, Intermountain Fire Sciences Laboratory, Missoula, MT).

surface and that introduced into the convection column along the trajectory from the surface to the level of sample collection, could affect the differential measurements, although ambient CO_2 is generally well-mixed in the boundary layer. However, the particle formation processes and effects of higher-than-ambient temperature conditions near the surface may affect particle formation, thus producing erroneous results. The processes described could have resulted in the outlier measurements of emission factors illustrated in Figs. 4.9 and 4.10.

Developing representative emission factors by weighting the measurements according to the carbon released during flaming and smoldering combustion can only be done using surface sampling and laboratory sampling techniques. Airborne sampling will be weighted according to the combustion conditions at the time of sample collection and by the rate of heat release providing the buoyancy required to loft the emissions to the height of emissions collection.

In Fig. 4.10, the emission factor response function for total PM (Ward and Hardy 1991) is lower than the data of Radke et al. (1991), based on measuring the size distribution of the particles and assuming spherical particles and a unit density. Ward and Hardy (1991) measured emission factors lower than those of White (1987) and Ward (1979). The southern pine fuels burned by White (1987) have a higher extractable resin content than other fuel types studied in the Pacific Northwest of the U.S. and the fuels studied in Brazil. The data of Ward (1979) are for the controlled combustion of α-pinene in a small reactor combustion chamber. One of the more
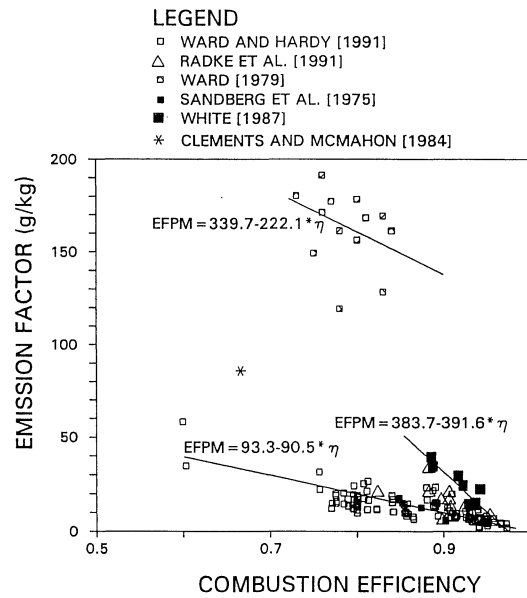
LEGEND
□ WARD AND HARDY [1991]
△ RADKE ET AL. [1991]
▫ WARD [1979]
■ SANDBERG ET AL. [1975]
▪ WHITE [1987]
* CLEMENTS AND MCMAHON [1984]

EFPM = 339.7-222.1*η

EFPM = 383.7-391.6*η

EFPM = 93.3-90.5*η

Figure 4.10 Emission factors for total particulate matter (without regard to size) for a range of combustion efficiency conditions in three different sets of combustion hood, ground, and airborne sampling, as compared to a controlled combustion apparatus with a model fuel of α-pinene. The linkages between fuel type, scale of combustion experiment, and systems for performing measurements need more research.

abundant oleoresin materials, α-pinene is found in pine needles in southeastern U.S.A. and was used as a model fuel for studying the controlled combustion of this class of fuels. The hypothesis for the work of Ward (1979) was that α-pinene or other terpenes with a similar basic structure to α-pinene would be evaporated from the pine needles in advance of the flames and could contribute to the advance of the flame envelope. It was hypothesized that the terpenes are responsible for a large fraction of the PM emissions. The results of White (1987) and Clements and McMahon (1984) for pine needle litter fuels of the southeastern U.S.A. provide some evidence in support of the hypothesis. The data point for thermoparticulates at η of 0.67 of Clements and McMahon (1984) fits an extension of the regression model of White (1987) (Fig. 4.10).

The measurements of Andreae et al. (1988) give additional credibility to the regression models developed by Ward and Hardy (1991). By calculating the EFPM value for a η of 0.908, a value of 12.2 g kg^{-1} is obtained. This falls within the range of EFPM values given by Andreae et al. (1988) for flight 16 in Brazil.

It is not clear whether PM emissions from open burning of biomass fuels are as well characterized as gaseous emissions or whether scaling factors for relating results from different sampling techniques are adequate (Figs. 4.9 and 4.10).

CONCLUSIONS AND FUTURE RESEARCH NEEDS

- Whereas CO and CH_4, for example, are linearly correlated, the ratio of CO/CO_2 is nonlinearly correlated with both CO and CH_4 and many of the other HC. In general, combustion efficiency (η) and modified combustion efficiency ($\hat{\eta}$) should be used as an independent variable in preference to CO/CO_2.
- CO_2 has been used to expand emission factor data for global estimates. However, combustion conditions that decrease the production of products of incomplete combustion tend to increase the production of CO_2. Serious errors may result from using a constant ratio.
- Although the main source of NO is from the oxidation of fuel-N, a second source of NO may result from the disassociation of molecular nitrogen under ideal burning conditions for very high-intensity fires.
- Progress has been made in the area of understanding the effects of cloud processing on smoke emissions; more work is needed, especially on the effects of water condensation processes on particles. The selective removal of both particles and gases needs further study.
- The electrical properties of aerosols may be of critical importance in understanding the life cycle of carbon particles in the atmosphere.
- The chemical composition of biomass is known to effect the release of trace emissions of nitrogen; however, little is known regarding other trace emissions either because the trace material may serve to catalyze the formation of products or lead to the formation of trace gases and particles.
- Although η and $\hat{\eta}$ work well as integrators of fuel morphology and weather influences on fire behavior and resulting emissions, variables more closely coupled to parameters used for characterizing ecosystems are needed to evaluate emissions and combustion factors.
- In an effort to develop protocols for evaluating emissions based on a few measurements of stable products of combustion, more emphasis needs to be placed on cross-correlating stable compounds with those that are reactive or are difficult to sample and characterize under field conditions. Field experiments need to be coupled with closely controlled and designed laboratory-scale experiments to gain an understanding of the mechanisms affecting the release of trace emissions.
- A new generation of techniques is needed for gas measurements similar to the FTIR system for quantifying emissions *in situ* rather than sampling, derivatization, or other difficult protocols. For PM or lidar, other nonintrusive remotely operated systems are needed for measuring the vertical and/or horizontal profiles.
- New methods for characterizing plume particle size distribution, shape of particles in smoke plumes, and the light extinction, scattering, and absorption characteristics of the particles are needed. Consideration needs to be given to the effect of wet atmospheres (where the relative humidity exceeds 60%–95%) on properties of particles affecting the absorption and scattering of light.

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