LABORATORY STUDIES OF SMOKE PROPERTIES AT NIST PAST, PRESENT, AND FUTURE[1]

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ABSTRACT

Burn facilities have been developed at NIST to measure the yield and optical properties of smoke at two fire scales. The small scale apparatus is used for burning samples about 10 cm in diameter (soup bowl size) and an intermediate scale apparatus is used for objects up to about 70 cm in diameter (bird bath size). Each of these facilities monitor the mass loss rate of the fuel with a load cell, the heat release rate by oxygen consumption calorimetry, mass concentration of smoke particulate with a filter, laser transmittance through the smoke, and the concentration of the major combustion gases. For the small scale apparatus, the radiant flux to the sample can be increased with a conical radiant heater and the composition of the air can be varied to study vitiated burning. Illustrative results for a variety of materials at the two scales are presented. Planned experiments involving vitiated burning and field scale measurements will be described.

Coupled with the intermediate scale facility is a specially designed aging and dilution chamber, which allows simulation of the smoke aging that occurs as a smoke plume rises in the atmosphere. Results on the effect of aging on the agglomerate size and optical properties are presented and compared with theory.

A transmission cell - reciprocal nephelometer has been developed to study the light scattering and absorption of smoke agglomerates. On-going efforts to quantify the instrument performance are discussed and recent results on the effect of cloud-processing on optical properties of smoke are presented.

1. INTRODUCTION

This paper describes laboratory scale facilities developed at NIST for characterizing the burning rate of liquid and solid fuels and properties of the resulting smoke. Before describing these facilities and some of the experimental results, a general perspective is provided regarding the effects of fire phenomena on the visibility of an object through a combustion generated smoke cloud. In the case of fire research directed at civilian safety, the basic issue is the visibility of an exit sign or a door in a smoke filled enclosure. A radiative transfer model for visibility including the effects of background lighting, the smoke distribution, the optical properties of the smoke, and multiple scattering has not been developed by the fire community. Still, there is a body of information in the fire research community that could be useful to the Army as input parameters for modeling target visibility through fire generated smoke.

[1] This research has been funded in part by Defense Nuclear Agency with Major Richard Hartley serving as scientific officer.

The burning rate of the fuel, the chemistry of the fuel, and the availability of oxygen for burning control the smoke emission (Mulholland et al., 1989 and Dod et al., 1989). The smoke emission is a key factor affecting visibility. The effect of the fire scale on the burning rate and smoke emission is an active area of research in the fire community. Fuels being studied include liquid hydrocarbons such as JP-4 and crude oil, wood, and plastics.

The spatial distribution of the smoke as it rises from the source will be strongly effected by the heat release rate of the fire, but it will also be affected by the wind velocity and the local topography. The modeling and characterization of fire plumes has in the past focused on the near field; that is, the plume has been characterized up to a height equal to several times the visible flame height. Because of recent interest in the environmental and climatic impact of fires, there are now several efforts in the fire research community including one at NIST by H. Baum (Evans et al., 1989) to develop a large eddy simulation of a wind blown plume. This information on both the concentration and distribution of the smoke is required for a full radiative transport model of visibility.

There is a data bank (Bankston et al., 1981; Seader and Einhorn, 1977) on the specific extinction coefficients of smoke obtained by the fire research community, though it is primarily limited to visible wavelengths. The earlier data were for small sample sizes, on the order of 10 cm length, but more recent experiments performed at NIST and elsewhere extended the laboratory scale measurements to burning objects on the order of 0.5 to 1 meter for a variety of fuels. For selected fuels including JP-4 and vegetative fuels, field scale burns involving 30 meter pool fires in the first case and more than 100 hectares slash fires in the latter case have been carried out.

Accurate measurement techniques are just being developed for routinely measuring the specific absorption and scattering coefficient of smoke. One of these instruments, the transmission cell - reciprocal nephelometer, is being developed at NIST to provide state of the art measurement accuracy. In addition to optical data on the smoke leaving the flame, there is also data for liquid fuels on the infrared radiant emission, which primarily arises from the near black-body radiation of the smoke in the flame. This information may be useful to the Army in regard to detection of fires and to the effect on target visibility. For example, the visibility of an object as viewed by an infrared scanner may be greatly reduced by the presence of an intense infrared emitter.

The optical theory of fractal smoke (Berry and Percival, 1986) is being used by a number of fire research investigators for interpreting optical measurements at visible wavelengths. There is a great potential for using this theory to model the optical properties of smoke over the much greater wavelength region of interest to the Army (visible to microwave wavelengths).

Before describing the facilities developed at NIST for measuring burning rate, smoke emission and smoke properties, there is a brief section defining the terminology used in the paper.

2. DEFINITION OF SMOKE PROPERTIES

The most rudimentary smoke quantity is the smoke yield, ϵ, which is defined as the mass of smoke aerosol generated per mass of fuel consumed. This quantity can be determined by a flux method or by a carbon balance method. The flux method simply consists of measuring the smoke collected on a filter, m_s; the mass loss of the sample, m_f; and the ratio of the mass flow of air through the exhaust duct to the mass flow through the filter sampler, ϕ. The smoke yield obtained by the flux method is denoted by ϵ_1.

$$\epsilon_1 = (m_s/m_f)\phi \tag{1}$$

The carbon balance method involves the determination of Y_s, the carbon mass in the smoke aerosol, as a fraction of the carbon mass in the total combustion products (CO_2, CO, and smoke aerosols). These represent the major carbon containing products of combustion for overventilated combustion. The contribution of unburned gaseous hydrocarbons to the carbon balance was of order 2% or less based on selected tests. By this method, the smoke yield, ϵ_2, is obtained as the product of Y_s and the mass fraction of carbon in the fuel, F_c.

$$\epsilon_2 = Y_s\ F_c . \tag{2}$$

Two specific light extinction areas are measured. The light extinction coefficient k is related to the light transmittance, I/I_o, via Bouguer's law,

$$I/I_o = e^{-kL}, \tag{3}$$

where L is the path length in meters. The specific extinction area on a fuel-pyrolysates basis, σ_f, is defined by

$$\sigma_f = k/(\dot{m}_f/\dot{V}) \ , \tag{4}$$

where $\dot{m}_f$ is the fuel mass loss rate and $\dot{V}$ is the volumetric flow rate of the combustion products through the exhaust duct. As a heuristic example, a value of 1 m^2/g for σ_f means that if the smoke produced by one gram of fuel were collected over a 1 m^2 area, the light incident on this area would be reduced by a factor of e.

The specific extinction area on a smoke particulate mass basis, σ_s, is defined by

$$\sigma_s = k/M_s , \tag{5}$$

where M_s is the mass concentration of the smoke where the transmittance measurement is being made. The quantity σ_s is an intrinsic property of the smoke depending on the wavelength of light, the refractive index of the smoke, and on the size and structure of the smoke particulate. The quantities σ_f and σ_s are related through the equation

$$\sigma_f = \epsilon\ \sigma_s . \tag{6}$$

The specific extinction area is equal to the sum of the scattering area, σ(scat), and the absorption area, σ(abs).

$$\sigma_s = \sigma(\text{scat}) + \sigma(\text{abs}) \tag{7}$$

The transmission cell - reciprocal nephelometer discussed below measures σ_s and σ(scat) simultaneously.

3. SMOKE COLLECTION FACILITIES

3.1 CONE CALORIMETER

The Cone Calorimeter was developed by Babrauskas (1982) to measure the heat release rate and related combustion properties of materials as a function of radiant flux. The conical radiant source (see Fig. 1) provides a uniform radiant flux over the 0.01 m² sample for fluxes up to 100 kW/m². After a warmup period for the radiant source to reach steady state, the sample is inserted and ignited via spark. The smoke aerosol and combustion gases rise, pass through the opening in the conical heater (for specimens in the horizontal orientation), then through a mixing orifice into the exhaust hood, and are finally sampled from a horizontal section of pipe, as indicated in figure 1. The fuel burning rate is monitored with a load cell; the heat release rate of the fuel is determined from monitoring the O_2 consumption in the exhaust gases (Huggett, 1980). The gases monitored include CO, CO_2, O_2, H_2O, HCℓ, and total hydrocarbons.

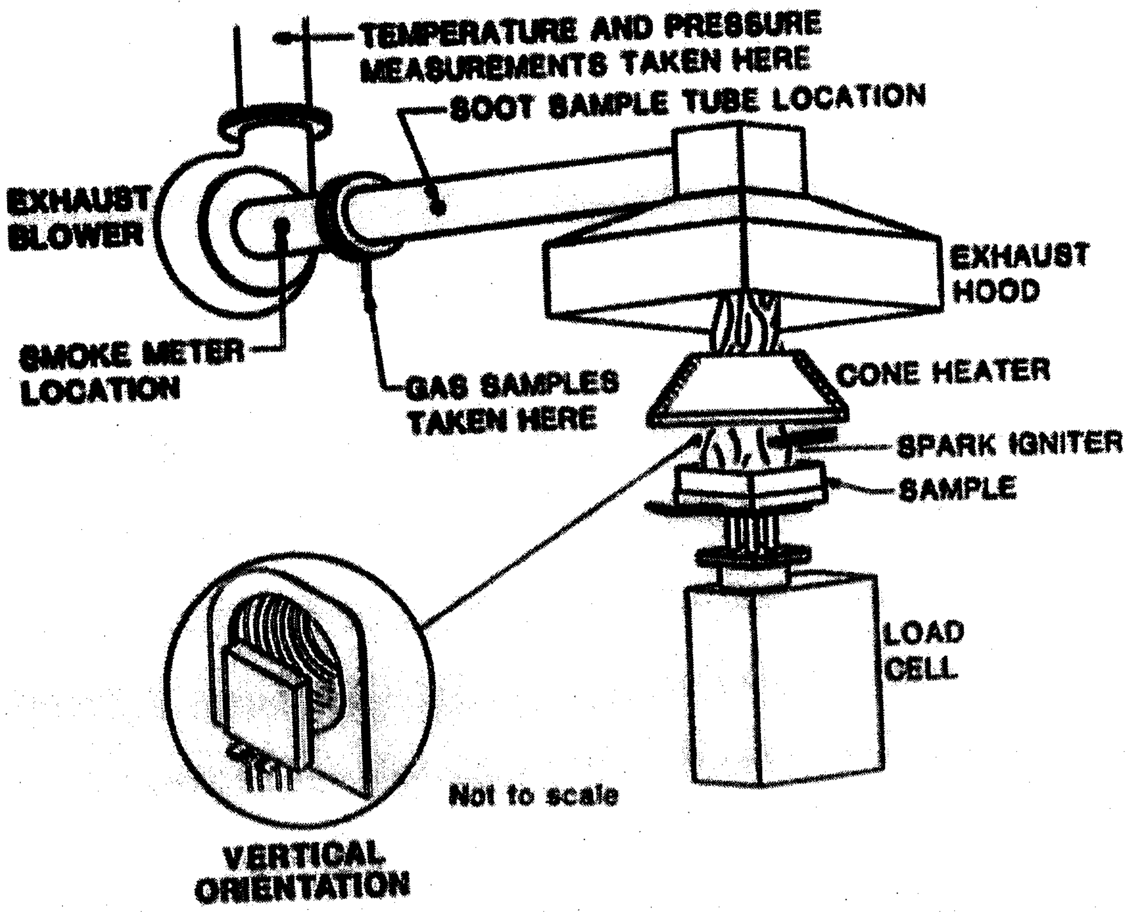

Figure 1. Small scale test facility. (After Mulholland et al., 1989)

The smoke particulate is collected on a Pallflex[2] 47 mm diameter fiber glass filter coated with polytetrafluoroethylene (PTFE). The flow through the filter system is servo-controlled to maintain a fixed-fraction mass flow rate relative to the main duct flow rate.

The optical extinction measurement is based on a He-Ne laser, λ=633 nm, with two silicon photodiodes, one for monitoring the laser intensity and the second to monitor the transmitted intensity. This system compensates for any variation in the laser intensity. This compensation is crucial to applying the instrument to weakly smoking fuels because of the small optical path length, about 0.11 m, and the normal drift in the laser output. Other important design features include a rigid mounting isolated from the fan vibrations and the use of purge air to minimize smoke deposition on the optical components.

3.2 INTERMEDIATE SCALE TEST FACILITY

This facility accommodates fire sizes up to about 400 kW, which corresponds to about a 0.6 m heptane pool fire. The fires are situated under a 2.4 m x 2.4 m collection hood (Fig. 2) with an adjustable exhaust rate up to about 2 m^3/s (4000 ft^3/min). A "tripper" orifice plate with a 0.45 m diameter is located at the base of the exhaust duct, 0.49 m diameter, to insure good mixing five duct diameters downstream at the sampling point. The mass loss rate of the burning fuel is monitored with a water cooled load cell with a sensitivity of about three grams. The heat release rate is determined from oxygen consumption calorimetry, as is done in the Cone Calorimeter; CO and CO_2 are measured in order to apply the carbon balance method to determine the smoke emission.

The filter collection system allows for the sequential collection of three filter samples over the course of a test. The transfer line, manifold, and filter holders are all heated to approximately match the stack temperature during the burn. This is done to minimize the evaporation/condensation of the smoke aerosol during transport or on the filter and to minimize the thermophoretic deposition of particles on the walls. The sample flow, about 10 L/min, and the nozzle inlet, 4.8 mm, were selected to ensure isokinetic sampling. The all-glass construction of the filter collection system allowed ready inspection of deposition and ease in cleaning. The major deposits are found to be at the two bends. Before each experiment, the heated filter system is leak-tested by attaching a dry test meter to the sampling probe.

The extinction measurement is obtained using essentially the same optics and electronics as in the Cone Calorimeter. The only conceptual difference is an 0.48 m pathlength, compared to a 0.11 m pathlength in the Cone Calorimeter. A slightly different mechanical mounting arrangement, involving a supporting ring structure, needed to be evolved to properly support the optics over the longer distance. Because of the longer pathlength, however, the sensitivity of the extinction measurement is greater for the intermediate scale apparatus.

[2] Certain commercial equipment, instruments, and materials are identified in order to adequately specify the experimental procedure. Such identification does not imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best for the purpose.

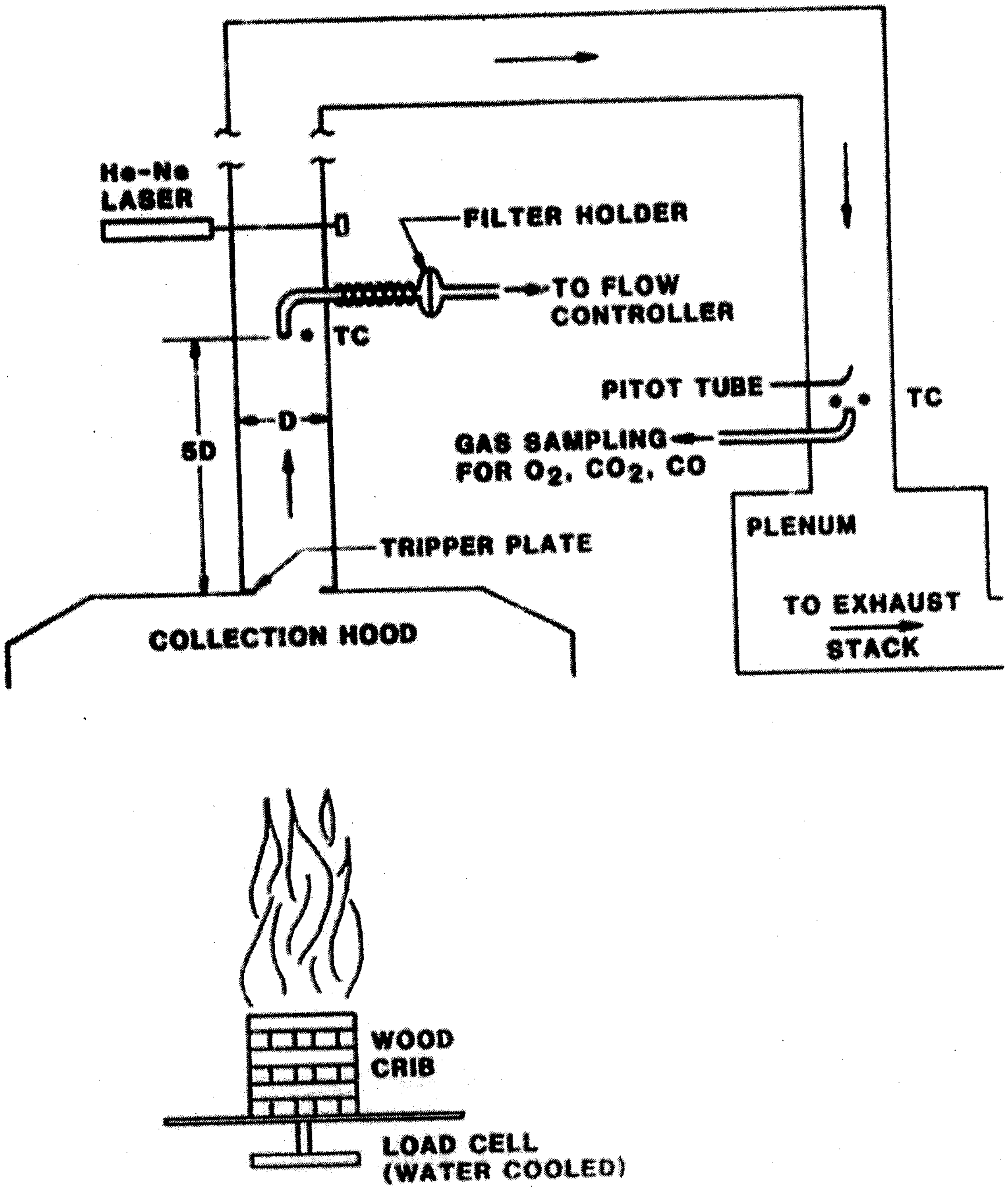

Figure 2. Intermediate scale smoke facility. (After Mulholland et al., 1989)

3.3 RESULTS FROM SMALL AND INTERMEDIATE SCALE FACILITIES

As an illustrative example, the specific extinction area for the smoke produced by burning a 10 cm by 10 cm by 2.5 cm thick sample of red oak in the Cone Calorimeter is plotted in Figure 3. Both the smoke yield (see Table 1) and the specific extinction area increased by almost a decade with increasing radiant flux. The major peaks in σ_f mirror the same peaks seen in the burning rate. The enhanced burning at the later stage results from reduced conductive heat loss when the thermal "wave" propagates through the entire sample. The values of ϵ and σ_f for wood burned in a horizontal configuration are approximately twice as large as those burned in a vertical configuration. The values in Table 1 correspond to an average of horizontal and vertical results.

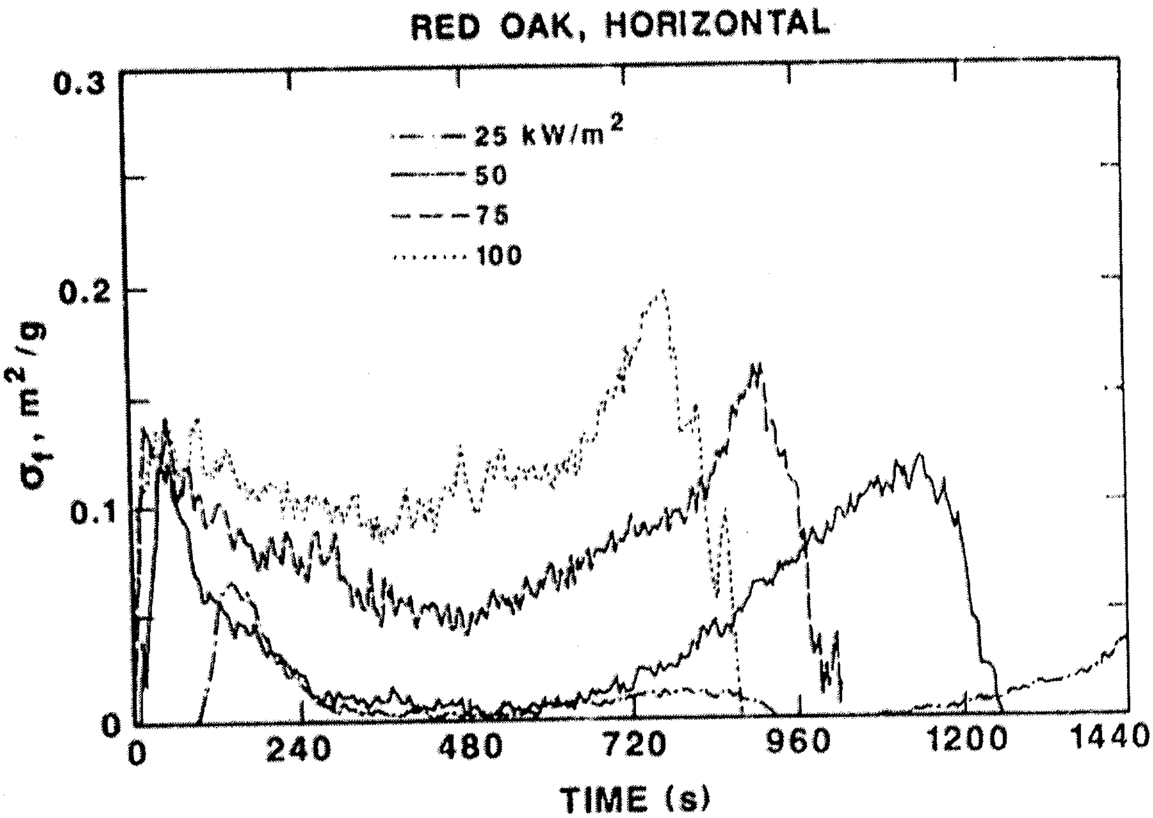

Figure 3. Specific extinction area, σ_f, vs. time for a sample of red oak lumber burning in the horizontal configuration. (After Mulholland et al., 1989)

For the intermediate scale burn, the wood was cut into sticks and assembled into layered "crib" structures. The value of ϵ for the larger scale tests, 0.004, is similar to the Cone Calorimeter result, 0.003, for comparable specific burning rate, m_f'' (indicated by bold print in Table 1). Results for heptane, ϵ = 0.012, and crude oil, ϵ = 0.085, given in Table 1, indicate the sensitivity of the smoke yield to the chemistry of the fuel. The mean large scale results agree with the small scale results for the comparable specific burning rate: heptane 0.011 vs 0.013, Prudhoe Bay crude oil 0.088 vs 0.090, and wood 0.004 vs 0.003. The agreement between large and small scale tests for σ_f is similar.

TABLE 1. COMPARISON OF SMALL AND INTERMEDIATE SCALE SMOKE PROPERTIES

Fuel/Conditions	Irrad. (kW/m^2)	$\dot{Q}$ (kW)	$\dot{m}_f''$ ($g/m^2 \cdot s$)	Comb. Eff.	ϵ	σ_f (m^2/g)	σ_s (m^2/g)
Heptane							
Large Scale							
310 mm pool		70	25	0.89	0.009	0.07	7
500 mm pool		240	28	0.94	0.012	0.10	8
Small Scale							
85 mm pool	0	3	10	0.99	0.010	0.06	8
	10	7	24	0.94	0.013	0.08	7
	20	10	35	0.97	0.010	0.07	8
	30	15	58	0.98	0.006	0.05	7
60 mm pool	0	1	9		0.015	0.15	10
	10	3	18		0.016	0.14	9
	20	5	38		0.013	0.12	9
	30	7	59		0.013	0.12	9
Crude oil							
Large Scale							
400 mm pool		65	14	34[a]	0.090	0.96	9.5
600 mm pool		185	(18)		0.085	-	8.7
Small Scale							
85 mm pool	0	1	5	41	0.098	1.06	11.7
	25	2	11	38	0.096	1.01	10.8
	40	4	18	37	0.083	1.00	12.5
	50	5	24	36	0.084	0.98	11.7
Wood							
Large Scale							
sugar pine							
1 crib		56	9[b]	0.66	0.004	0.03	9
3 cribs		254	13	0.69	0.004	0.04	9
Small Scale							
Red oak, 100 mm	25	1	9	0.55	0.002	0.02	11
	50	1	12	0.56	0.004	0.04	11
	75	2	15	0.56	0.006	0.07	13
	100	2	19	0.60	0.011	0.09	10

a.. The heat of combustion in MJ/kg.

b.. The effective surface area for combustion is taken as half the total surface area of all the individual sticks.

The average value of the specific extinction area, σ_s, for each fuel is in the range 8 to 12 m^2/g for the Cone Calorimeter data, while for the large scale tests the range is less, 8 to 9 m^2/g. The thermophoretic deposition in the Cone Calorimeter sampling probe is possibly responsible for the larger variability in σ_s for the Cone. The fact that σ_s is independent of the fuel burning is a key result of our study.

Currently, the effect of vitiated air on the combustion products including smoke particulate is being studied using a Cone Calorimeter with an enclosed burn area. Measurements are being made at oxygen concentration down to the flammable limit, on the order of 12 to 15% depending on the fuel.

A major study is underway to characterize the smoke yield of burning crude oil at large scale. Preliminary measurements performed by Mulholland and Koseki at the Fire Research Institute in Japan indicate that the smoke yield for crude oil increased by about a factor of two as the fire scale was increased from 0.6 m to about 3 m diameter pools. Plans are being made to perform field experiments with crude oil for pool diameters up to about 100 meters with support from the Mineral Management Service.

4. AGING AND DILUTION CHAMBER

In a smoke plume, the optical and physical properties of smoke change as the smoke ages. Close to the fire source, these changes occur in hot, fresh smoke, but downstream in the plume, changes also occur in the cooler smoke which may or may not be diluted. In order to study how these changes affect properties, such as the light extinction and size distribution of the smoke particles, an Aging and Dilution chamber has been fabricated (Evans et al., 1989).

The chamber illustrated in Figure 4 is a 1 m^3 aluminum box which has been lined with stainless steel to reduce corrosion from the hot combustion gases. Forty-eight mica resistance strip heaters are attached to the aluminum walls which evenly distribute the heat for wall temperatures up to 150 °C. The intake ductwork and valve can also be electrically heated. For experiments which require cooler or more dilute smoke, dilution air can be added via a port in the intake line. The port is designed to allow adequate mixing of the dilution air and smoke before entering the chamber. Orifice flowmeters permit measurement of the inlet, exhaust and dilution air flow rates. To allow for rapid filling of the chamber, a variable speed exhaust fan pulls smoke through the chamber at flow rates up to 8 m^3 per minute. Using the variable speed fan and inlet tips of diameters ranging from 1.5 to 10 cm, the inlet velocity can be matched with the stack velocity for isokinetic sampling. The smoke enters and exits the chamber through 10 cm diameter stainless steel ductwork. After the chamber is filled and well mixed, two stainless steel butterfly valves, one on the inlet and one on the outlet, are simultaneously closed to capture a 1 m^3 sample. A 100 liter piston which is located on top of the chamber allows the volume of the chamber to be reduced as samples of smoke are withdrawn from any of the 20 sampling ports. A pair of the sampling ports have been adapted for a three wavelength photometer for extinction measurements. Some of the other instruments used with the chamber include the tapered element oscillating microbalance, a seven stage cascade impactor, a condensation nuclei counter and the transmission cell-reciprocal nephelometer.

In a preliminary study with crude oil fires, light extinction, mass concentration, and number concentration were measured with a photometer, microbalance, and nuclei counter. The photometer, which is similar to the design by Cashdollar et al. (1979), consists of a white light source, two beam splitters, three interference filters and three photodiodes. Mounted on opposite walls of the chamber, the light source and the receiving optics are separated by pathlength of 1.1 meters. The light extinction coefficient at 450, 630 and 1000 nm is determined from the per cent transmission at each wavelength. In order to calculate the extinction coefficient per unit mass concentration of smoke, σ_s, the mass concentration of the smoke in the chamber is determined with the Tâpered Element Oscillating Microbalance (TEOM). Approximately two liters of smoke are necessary to make a mass concentration measurement.

A series of experiments were performed to look at the effect of aging on smoke properties. Samples of crude oil smoke were collected and aged for up to 150 minutes, during which time the concentration and light extinction measurements were obtained. The mass concentration peaked at about 200 mg/m^3 just after the chamber was filled with smoke. Generally, the mass concentration decreased by a factor of about 2 over the 90 minute aging period while the number concentration decreased by about a factor of 60 implying a 30 fold increase in the mass per smoke agglomerate as a result of agglomeration of particles. Even though there was this large increase in agglomerate mass, the specific extinction area, σ_s, at 450, 630 and 1000 nm did not change significantly over a 90 minute aging experiment (Figure 5). Three wavelength extinction data were collected every 10 minutes throughout the aging period. The mean value for σ_s were 5.1 (1000 nm), 7.8 (630 nm), and 9.7 (450 nm). The independence of the specific extinction area to aging is consistent with the optics of fractal agglomerate (Berry and Percival, 1986 and Nelson, 1989).

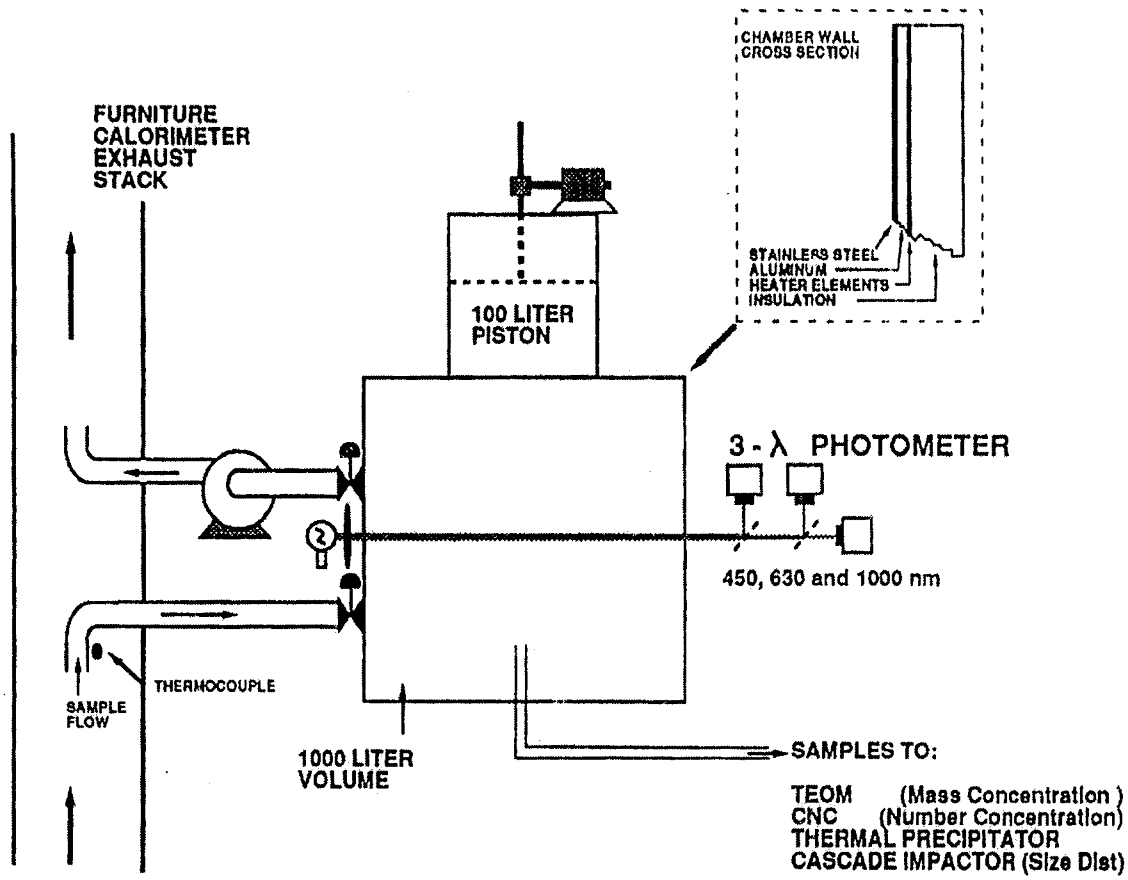

Figure 4. Smoke Aging and Dilution Chamber (CNC - Condensation Nucleus Counter, TEOM - Tapered Element Oscillating Microbalance), (After Evans et al., 1989)

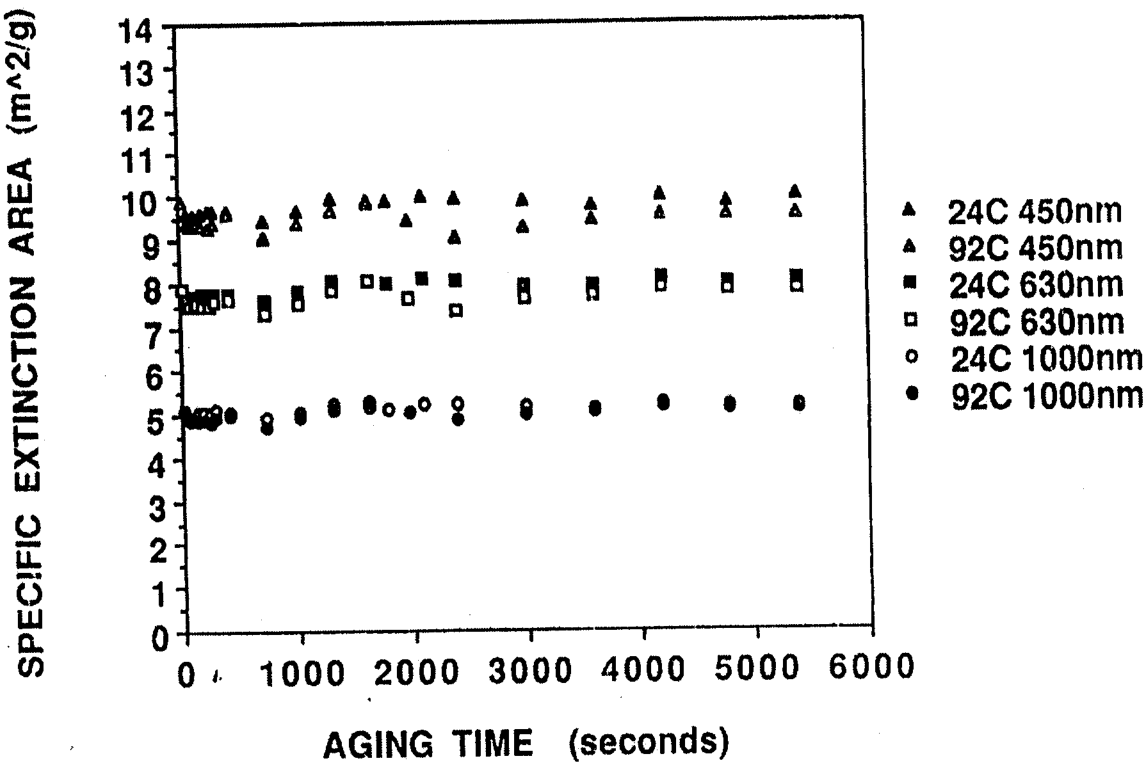

Figure 5. The specific extinction area, σ_s, at 450 nm, 630 nm, and 1000 nm vs. time for aging of crude oil smoke at ambient and elevated temperature. (After Evans et al., 1990)

5. TRANSMISSION CELL - RECIPROCAL NEPHELOMETER

A one meter path length transmission cell-reciprocal nephelometer (TCRN) similar to Gerber's design (1982) has been developed to measure simultaneously the extinction and total scattering cross sections of the smoke. A schematic of the instrument design is shown in Figure 6. The novel feature of the instrument is the use of a cosine sensor to collect light scattered along the 1 meter pathlength with scattering angles between 5°and 175°. As shown by Gerber (1982), the cosine sensor yields an output proportional to the total scattering intensity over the angle range from 5° to 175°. Other design features include measurement of percent transmission at a sensitivity of 0.03% by monitoring the intensity of both the incident and transmitted beam, purge air on windows to minimize smoke deposition, and absolute calibration of scattering coefficient by measuring the extinction coefficient for a non-absorbing aerosol. A unique capability of this instrument is the measurement of the absorption cross section of the aerosol by taking the difference between the extinction and total scattering. We are not aware of any other instrument capable of performing an accurate measurement of the specific absorption coefficient.

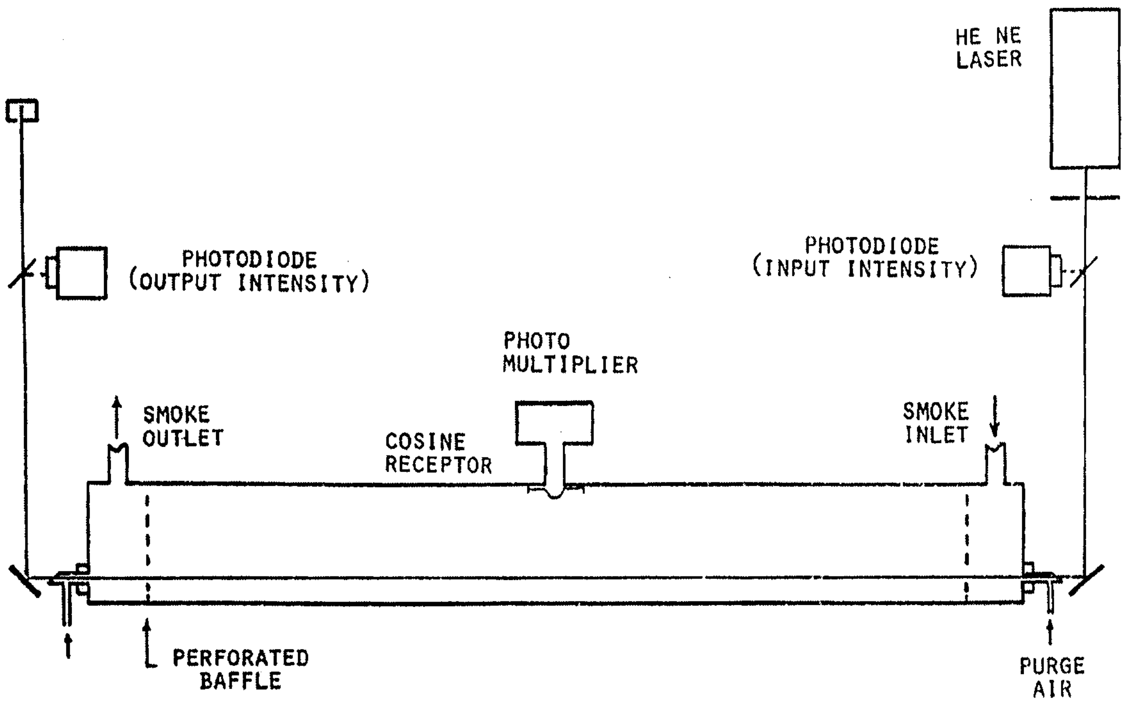

Figure 6. Schematic diagram of the Transmission Cell - Reciprocal Nephelometer.

Preliminary results have been obtained for the optical properties of "fresh" and "cloud-processed smoke." Cloud-like conditions are likely to occur above a large fire and it is of interest to know the effect of these conditions on the optical properties of the smoke. The smoke was produced by burning acetylene with a laminar diffusion burner. A Pollak type expansion cloud chamber (dimensions scaled up to 9 liter, 7.5 cm diameter) fabricated at Desert Research Institute was filled with smoke and pressurized to 2.1×10^4 Pa above ambient. A sudden expansion produced a cloud of water droplets with the smoke particles acting as nuclei. The droplets were evaporated, and the "processed" smoke was passed into the 9 liter TCRN. The light transmission through the cell, the total scattering (both monitored at λ=633 nm), and the mass concentration of the smoke by an oscillatory microbalance were measured simultaneously as the cloud-processed smoke moved through the cell.

As indicated in Table 2, the specific scattering area and the single scattering albedo, ω, defined as the ratio of the scattering area to the extinction area, increased by about a factor of two as a result of the cloud-processing. Electron micrograph of the cloud processed soot indicates that the low density agglomerate structure of the fresh soot has been compacted by the cloud-processing. The increase in scattering for this soot with a mass averaged cluster size of 420 spherules with diameter of about 30 nm is consistent with the Berry - Percival Theory of the optics of fractal smoke (1986).

TABLE 2. OPTICAL PROPERTIES OF FRESH AND CLOUD-PROCESSED ACETYLENE SMOKE

Type of Smoke	σ_s m^2/g	σ(scat) m^2/g	σ(abs) m^2/g	ω
Fresh	9.1±0.4	2.2±0.1	6.9±0.2	0.24±0.01
$2.1X10^4$ Pa above ambient/130%[a]	10.8±0.8	4.0±0.7	6.7±0.7	0.37±0.06

[a] Estimated maximum supersaturation.

A detailed radiative transport model is being developed for the transmission cell - reciprocal nephelometer to include both the finite size effect of the cosine sensor and its position dependent transmission coefficient. This will lead to the state of the art accuracy in the measurement of single scattering albedo and specific absorption area.

6. CONCLUSION

Unique laboratory scale facilities have been developed for studying the burning rate of fuels and the smoke properties including the smoke yield and the optical properties of fresh, aged, and cloud-processed smoke. Major results include:

1. While the smoke yield has been shown to be sensitive to fuel type and burning condition, a near universal specific extinction coefficient, σ_s, of 8-9 m^2/g at 630 nm is obtained independent of the fuel type.

2. It has been shown that aging of crude oil smoke to the point where the average agglomerate mass has increased 30 fold has little effect on the specific extinction coefficient.

3. The structure and optical properties of smoke are changed by cloud-processing.

4. The optical theory of fractal agglomerates developed by Berry and Percival (1986) and Nelson (1989) is consistent with the measured effects of aging and cloud-processing on the optical properties of smoke.

ACKNOWLEDGMENT

The cloud-processing experiment was carried out in collaboration with J. Hallett, J. Hudson, and F. Rogers from Desert Research Institute.

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