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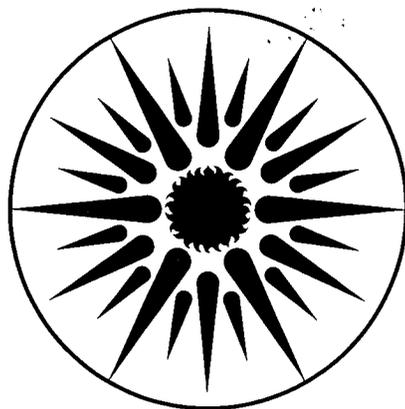
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SMOKE EMISSION FACTORS FROM MEDIUM SCALE FIRES: PART 2*

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ABSTRACT

Smoke emission factors, (i.e., the mass of smoke per mass of fuel burned), were measured in eleven separate experiments. The size distribution of the smoke particles was determined using a cascade impactor. The percentages of "black" carbon (also called "graphitic" or "elemental" carbon) and organic carbon have been determined for all the experiments as a function of particle aerodynamic diameter. Values in the range of .1 to .2% are reported for the smoke particle emission factors for Douglas fir whole wood and plywood burning under well ventilated conditions. Approximately 65% of the particles have aerodynamic diameters less than 1 μm . Douglas fir whole wood gave smoke emission factors in the range of 2 to 3.5% when burned under poorly ventilated conditions representative of a building fire that is limited by air entrainment. For this case the size distribution was much broader, with substantial quantities of particles up to 5 μm aerodynamic diameter. For all experiments, the black carbon content represented between 50 and 75% of the total mass of the smoke particles. The smoke emission factor for burning asphalt roofing shingles is reported as 12.1% with a black carbon content greater than 70%. Over half of the mass consisted of particles of less than 1 μm aerodynamic diameter.

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INTRODUCTION

Previous measurements of smoke emission factors have focussed primarily upon plume opacity (Rashash & Pratt, 1979) or upon the integrated mass of particle emissions (Mulholland et al., 1988; Patterson & McMahan, 1984). Accurate prediction through modelling of the effects of post-nuclear exchange smoke requires that emission factors, optical properties of smokes and rainout/washout probabilities be known (Penner, 1987; Turco et al., 1987). Particle size and composition affect both optical properties and wet removal probabilities.

In an earlier study, we performed a series of bench scale fire experiments to measure the smoke emission factors as a function of smoke particle size for four materials (Douglas fir plywood, Douglas fir whole wood, birch whole wood, and flexible polyurethane foam), which would be common in an urban fire (Dod et al., 1985). Smoke samples were fractionated by size using a cascade impactor, and the mass of particles in each size range was determined. The smoke emission factors measured in our bench scale study (except for Douglas fir whole wood) were approximately half an order of magnitude below those used by two of the definitive reviews of Nuclear Winter (Turco et al., 1983; National Research Council, 1985).

It is known that smoke emission factors are significantly different for fires of different sizes (Turco et al., 1987). Extrapolation from bench scale experiments to fires encompassing an area of several to many square kilometers is unlikely to be accurate. Research fires approaching such large magnitudes are not possible. We report here measurements of particulate emission factors, size distributions and composition for a set of medium-scale (0.2-1.2 MW) fires using as fuels common urban building materials (e.g., plywood, whole wood and asphalt roofing shingles).

EXPERIMENTAL:

The configuration of the apparatus for the experiments is described in Part 1 (Brown et al., 1988) of this series of papers. The principal location for aerosol particulate sampling was in the exhaust duct at a point approximately 2 m downstream of the straightening vanes. Two 6.4 mm o.d. (3.0 mm i.d.) smoke particle sampling probes were inserted into the duct to accommodate the impactor and a stacked-filter pair. Each probe inlet was designed to provide isokinetic sampling (at 5.66 l/min) of the smoke stream in the duct (velocity 13 m/sec) at a distance of approximately 6 cm from the duct wall, a distance adequate to avoid effects of the wall on stream velocity. Each probe line was straight except for two 15 degree bends with a 13 mm radius which were necessary to extract the sample from the duct and connect it to the sampling device. Additional sampling was conducted on all but the first two experiments by placing a 6.4 mm o.d. tube (5.3 mm i.d.) into the plenum immediately above the hood and leading it 2 m upward to the roof. Since the flow velocities in the plenum are not well defined and significant particle concentration gradients are visually apparent, no attempt was made to establish isokinetic conditions at this probe inlet. The sample taken from the plenum, another stacked-filter pair, was used to provide qualitative information regarding changes in the particulate carbon composition which could potentially occur during the 0.3 to 0.5 sec transit time from the plenum to the primary sampling ports.

The duct has been shown to have a uniform velocity field except in the immediate vicinity of the wall. We have assumed that the smoke particle concentration in the duct is uniform over the area of uniform velocity. The agreement of emission factors calculated from the samples from the two probe lines indicates that the assumption is valid. Velocity variations will result

in small uncertainties in the total emission factors and should not affect the proportions of black carbon, organic carbon and non-carbon mass.

Experiments using asphalt roofing and solid wood (under ventilation restricted conditions) produced such heavy smoke that dilution was necessary to avoid clogging of the impactor. This was accomplished by inserting into the impactor probe line a chamber through which continuously filtered air was recirculated by a diaphragm pump at 51 l/min to provide the impactor with only 1/10 of the duct particle concentration. The decision whether to use the dilution chamber was made prior to each experiment, and was based upon expected smoke production. All flow rates were determined by using the pressure drops measured across orifices located near the pumps, and were manually controlled by valves located between the pumps and the orifices.

During the course of each experiment, the gas velocity in the duct showed some variation, while the sampling flow rates were maintained constant. The probe inlet was designed to be isokinetic at a duct velocity of 13 m/sec, whereas the observed velocity was in the range 9 m/sec to 13 m/sec. One consequence of this variation is a slight enhancement or deficiency in large particles observed in the samples. In practice this was insignificant because very low concentrations were observed for particles larger than approximately 5 μm . A more important consequence of the variation in duct air velocity is that, with a constant sampling flow rate, the fraction of the smoke sampled varies inversely to duct velocity. Particle emission factors were calculated using the average duct flow rate during the sampling period, and this necessary simplification produces an estimated uncertainty of <10% in the absolute value of emission factors determined.

The impactor used for these studies was a Pilat Mark 5 Source Test Cascade Impactor (Pollution Control Systems Corp.), with jet stages chosen to

provide loadings adequate for accurate analysis with reasonable size ranges. To be able to make the mass and carbon measurements on each stage, it was necessary to line the impaction plates with aluminum foil which had been sequentially sonicated in carbon tetrachloride and acetone. All filters used were 47 mm quartz fiber, which had been fired in air at 800°C for 4 to 6 hours to remove all combustible carbon. For all experiments, the stage 1 collection plate (>56 μm particles) was coated with a thin layer of silicone grease to minimize bounce and re-entrainment of large particles. For the same purpose, the stage 2 collection plate (25-56 μm particles) was also greased for some experiments. This treatment rendered these stages unusable for mass or carbon determination, but visual observation indicated that little of the sample mass was present on these plates.

It has been shown that hydrocarbon vapors are adsorbed on quartz fiber filters when used for ambient air sampling (Cadle et al., 1983). To correct for this effect in these experiments, a filter stack configuration using two stainless steel filter holders in series was used for collection. The second filter was used as a blank to account for hydrocarbon vapor condensation and was used to correct the mass and carbon determinations made on all other filter samples.

A Cahn Model 4700 electrobalance was used to determine sample mass for each filter and ungreased impactor collection foil. Each filter or foil was equilibrated prior to use for a minimum of 24 hrs in a chamber at 45% relative humidity, and then weighed to the nearest microgram. After collection, the samples (and appropriate blanks) were returned to the chamber, and the equilibration and weighing procedure was repeated.

Total carbon content was determined on each sample by combustion in oxygen, followed by detection of the resultant CO_2 with a Coulometrics, Inc.,

Model 5010 CO₂ coulometer (Huffman,1977). This system has been shown to have an accuracy of $\pm 2\%$ or 1 μg , whichever is greater. Portions of the filters and foils containing at least 20 μg of carbon were chosen for analysis when the loading allowed, yielding an error in individual determinations of no more than 5%.

The carbon content of the samples was further characterized by a thermal evolved gas analysis (EGA) for carbon. This technique is capable of characterizing collected aerosol carbon particles by type: organic, black, and carbonate carbon (Dod & Novakov,1982; Novakov,1981). All the samples analyzed in this study showed a dominant black carbon peak with no significant overlap interference, allowing for an unambiguous differentiation of carbon type. No mineral carbonate was seen in any sample.

Sampling procedure reliability can be ascertained from a comparison of the total particulate carbon emission factors determined from the duct total filter (front filter in the stack) and the sum of the carbon collected in the impactor (0-56 μm). For fires without ventilation restriction the agreement is good, with the sum of impactor carbon equal to $92 \pm 7\%$ of the carbon collected by the total filter. That the sampling devices agree to this extent indicates that few of the smoke particles are of such large diameter as to be intercepted by the greased (and unanalyzed) upper stages of the impactor. This agreement also implies that the particle concentration in the duct is uniform at the location of the probe inlets. This comparison was not applied to the ventilation restricted fires since particle loadings of the total filters were extremely heavy and non-uniform in nature, making selection of a representative sample for analysis not possible. However, good agreement in total mass between the two samplers was observed for the ventilation restricted experiments.

Error in mass determination, estimated from blank filters and foils, is $\pm 12 \mu\text{g}$ for all except impactor afterfilters. Total smoke particulate mass measured on the afterfilters was usually less than the carbon mass present on those filters. This anomaly results from the nature of the filter material and the conditions under which it is used. Under some circumstances, loss of fibers can be expected from a quartz mat which has had the organic carbon binder removed prior to use. Consequently, non-carbon mass for the $<0.28 \mu\text{m}$ particles is most probably underreported for each experiment. For total carbon determinations, the estimated errors for filters and foils are respectively 0.6 and 4 μg , with the exception of the those used for studies of underventilated fires.

RESULTS AND DISCUSSION

The smoke emission factors for the fires in this series of experiments are summarized in Table I, and the averages for each fuel type and combustion condition are shown in Figures 1 and 2. Fuel configurations are discussed in more detail in the companion paper. Experiments 1, 3, 4 and 5 each used parallel 1.2 m by 1.8 m sheets of 1.3 cm thick 5-ply CDX grade Douglas fir plywood with a 15 cm spacing. The measured particulate carbon emission factors for these experiments were nearly identical ($0.17 \pm 0.01\%$), indicating the degree of reproducibility which can be achieved for specimens burning under similar conditions. Experiment 2 used identical plywood sheets, but was configured with a 10 cm gap between them, causing the fire to have more limited ventilation than the others. This reduction in ventilation results in the large value for the measured emission factor. Experiment 10 utilized a 3- rather than 5-ply product and as such is not directly comparable, since it has thicker veneer layers and contains relatively less adhesive. Because of this construction, the fuel in experiment 10 may have burned in a manner more

similar to whole wood. As an indication of this, the measured emission factor for this experiment is intermediate between those of the other plywood fires and that measured for a Douglas fir whole-wood crib burning in the open (experiment 11). The character of the smoke particles produced in each of the plywood fires is very similar, each containing 65 to 70% black carbon by weight. (Experiment 1 appears not to be in this range, but if the total mass is estimated by adding an average of the amount of non-carbon mass found for the other plywood fires, this experiment is also consistent.)

As indicated above, when a Douglas fir whole wood crib was burned (experiment 11), an even lower emission factor (0.09%) is observed. The reason for this is due to differences in the combustion which results from differences in fuel geometry. These are discussed in our companion paper.

When wood cribs are burned under limited ventilation conditions in an enclosed room with a single window (0.76 by 1.0 m), an increase in smoke particle emission factors of greater than an order of magnitude is observed (experiments 12 and 13). Although the combustion characteristics of both these experiments are very reproducible, the particle collection period for each of these experiments is different, and this may account for the difference in measured emission factors.

Burning of asphalt roofing shingles produced the largest emission factors observed in this series of experiments; smoke particulates in excess of 10% of the consumed fuel mass were observed for both fires. Particle collection for experiment 9 was initiated at an earlier time and was continued for a period 50% longer than for experiment 24. As is apparent in Figure 2, the average emission factor is nearly two orders of magnitude greater than that observed for the wood fires burning under similar ventilation conditions.

9

For all experiments performed in this study, regardless of fuel type or ventilation conditions, greater than 70% of the particulate carbon was black carbon which corresponded to 55-77% of total smoke particle mass. This similarity in smoke composition for these experiments is likely to have resulted from the residence time - temperature history relationship which occurs for particles in fires of this magnitude.

Figures 3 through 6 show smoke particle size distribution functions in which a normalized mass function, $dM/Md(\log(D_p))$, is plotted versus the particle aerodynamic diameter (D_p) on a logarithmic scale. In such a plot, the area of each region under the curve corresponds to the relative mass. The lower particle size limit, $2 \times 10^{-2} \mu\text{m}$ is an arbitrary, although realistic, value.

Figure 3 shows the distribution function for smoke from a well-ventilated plywood experiment. The majority of the carbon is found in the finest particle fraction (impactor afterfilter), with only a relatively small percentage having aerodynamic diameters greater than $1 \mu\text{m}$. Figure 4 shows a similar distribution function for a well-ventilated solid wood crib. Although the emission factor for experiment 11 is lower than that of experiment 5 by a factor of two, the composition and size distribution of the particles is very similar for both experiments. The larger amount of non-carbon mass from the solid wood burning may be, in part, an artifact of mass determinations for lightly loaded samples.

When wood was burned in an underventilated environment (e.g., expt. 13), a large change was observed in the particle size distribution as well as in emission factor (Figure 5). Although no significant change in particle composition was observed, the median particle size shifted from $<0.2 \mu\text{m}$ to greater than $1 \mu\text{m}$, with measurable amounts of black carbon present in particles

in the 10 μ m range. While the order of magnitude increase in emission factor is most likely due to the limited ventilation (Turco et al., 1987; Brown et al., 1988), the increase in particle size may be a product of increased residence time in an environment of high particle concentration allowing a greater degree of coagulation. The product particles are of a size which would be more subject to removal by rainout or washout (Penner, 1987).

Smoke from the burning of asphalt roofing shingles (e.g., Expt. 9) is distributed across the entire size range measured (Figure 6). Visual observation and mass balance mentioned above showed few or no particles present in larger sizes. Particle composition, however, was very similar to that from the wood fires. Although the largest size fraction is present on the afterfilter (<0.3 μ m), there is an apparent peak in the fraction of about 1 μ m, similar to that of the underventilated wood fires. This provides support for the hypothesis that further coagulation during residence in a region of high particle concentration may be occurring, since the smoke from the roofing fires was very dense.

Analyses of filter samples collected from the plenum were compared to those collected from the exhaust duct, and no significant differences in smoke particle composition were seen, showing that no appreciable amount of organic vapors were deposited on the particles from that point to the primary sampling ports.

The morphology of the smoke particles produced in these experiments has been observed by both optical and electron microscopy. They have an open, lace-like appearance characteristic of soot particles which are largely black carbon. Further description of morphology will follow in a later paper.

CONCLUSIONS

Particulate emission factors from the medium scale burning of wood and asphalt roofing, under open as well as restricted ventilation conditions, varied from less than 0.1% to greater than 10%. The dominant fraction (60-75%) of the smoke particles for all experiments in this study was black carbon. The relative composition showed very little change with fuel type or burning environment.

The results from this series of experiments differ significantly from those which we obtained in bench scale experiments. In the earlier bench scale, study the smoke emission factor was determined to be 0.6% for Douglas fir plywood, 2.3% for Douglas fir whole wood, 0.3% for birch whole wood, and 0.5% the flexible polyurethane foam (Dod et al., 1985). The higher smoke production of the whole wood was probably due to smoldering which produced more unburned aerosols. The black carbon constituted 1% of the total particulate mass for Douglas fir whole wood, 13% for the plywood, 4% for the birch whole wood, and 66% for flexible polyurethane foam. With the exception of the plastic foam, these fractions are much below those observed in our current work. This is most probably due to the more vigorous burning present in the larger fires, or to the extended flame environment. The difference in character between emissions from bench and medium scale fires did not, however, change the size distributions; for well-ventilated wood fires, the mass median aerodynamic diameter is below 0.2 μm for both fire scales.

Experiments are in progress to extend this work to liquid fuels and to explore the effects of scaling, geometry and materials composition on emissions to the extent that our facility will allow.

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Table I. Smoke Emission Factors

EXPT.	FUEL	EMISSION FACTORS			TOTAL MASS
		NON- CARBON	CARBON		
			BLACK	ORGANIC	
1	plywood	N/A	.13%	.03%	.16%
2	plywood	.00%	.17%	.06%	.24%
3	plywood	.02%	.12%	.04%	.18%
4	plywood	.03%	.13%	.04%	.20%
5	plywood	.01%	.13%	.05%	.20%
9	asphalt shingles	2.05%	10.03%	1.85%	13.94%
10	plywood	.04%	.09%	.01%	.14%
11	wood crib in open	.01%	.05%	.02%	.09%
12	3 wood cribs in room	.53%	1.12%	.29%	1.93%
13	3 wood cribs in room	.31%	2.66%	.54%	3.51%
24	asphalt shingles	1.86%	6.23%	2.12%	10.22%

LIST OF FIGURE CAPTIONS

- Fig. 1 Smoke emission factors measured for burning wood under well-ventilated conditions. (Average of all experiments with each fuel type).
- Fig. 2 Smoke emission factors for all fuels and conditions. WOOD* indicates whole wood cribs burned under limited ventilation conditions. (Average of all experiments with each fuel type).
- Fig. 3 Distribution function for emissions from burning of plywood (Experiment 5).
- Fig. 4 Distribution function for emissions from burning of a whole wood crib with good ventilation (Experiment 11).
- Fig. 5 Distribution function for emissions from burning of whole wood cribs under limited ventilation conditions (Experiment 13).
- Fig. 6 Distribution function for emissions from burning of asphalt roofing shingles (Experiment 9).

Average particle emissions Ventilated wood fires

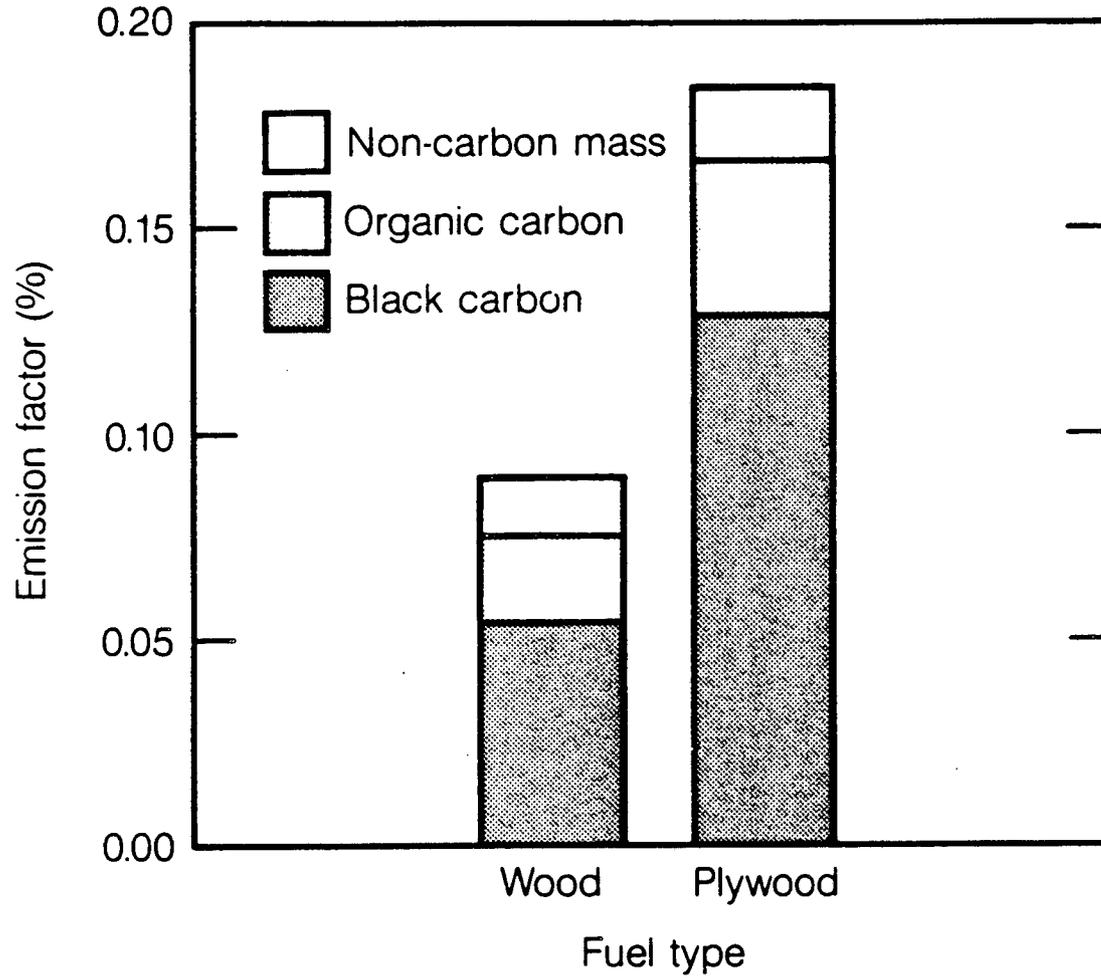


Figure 1

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Average particle emissions All fuel types

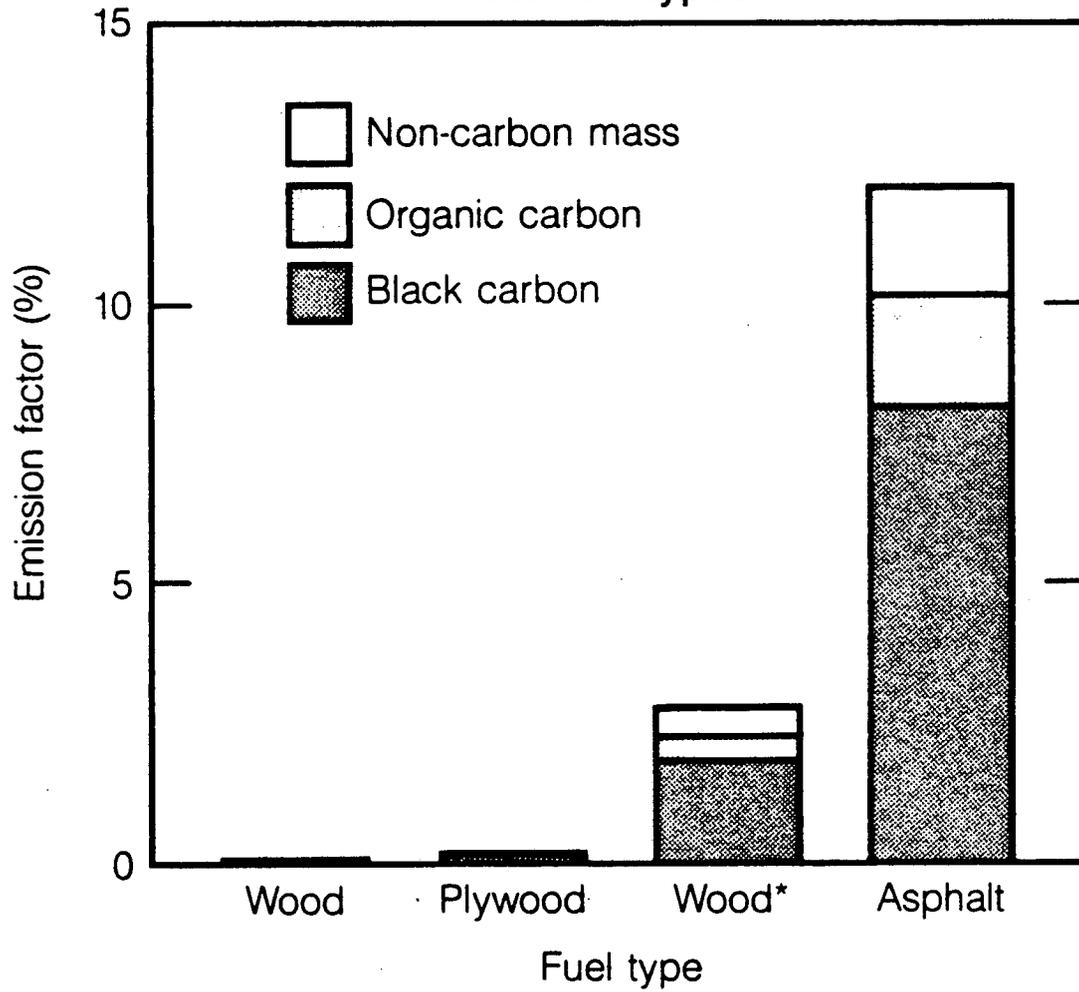


Figure 2

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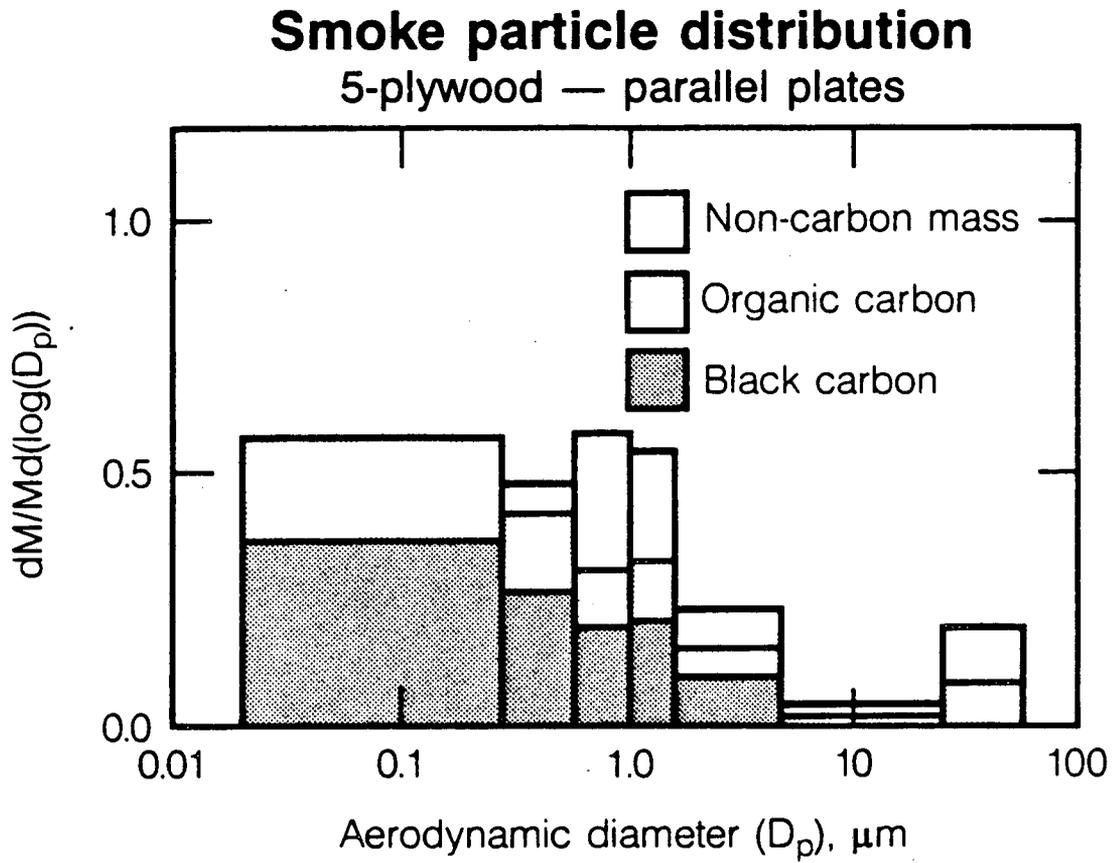


Figure 3

XBL 883-6017

Smoke particle distribution Well ventilated wood crib

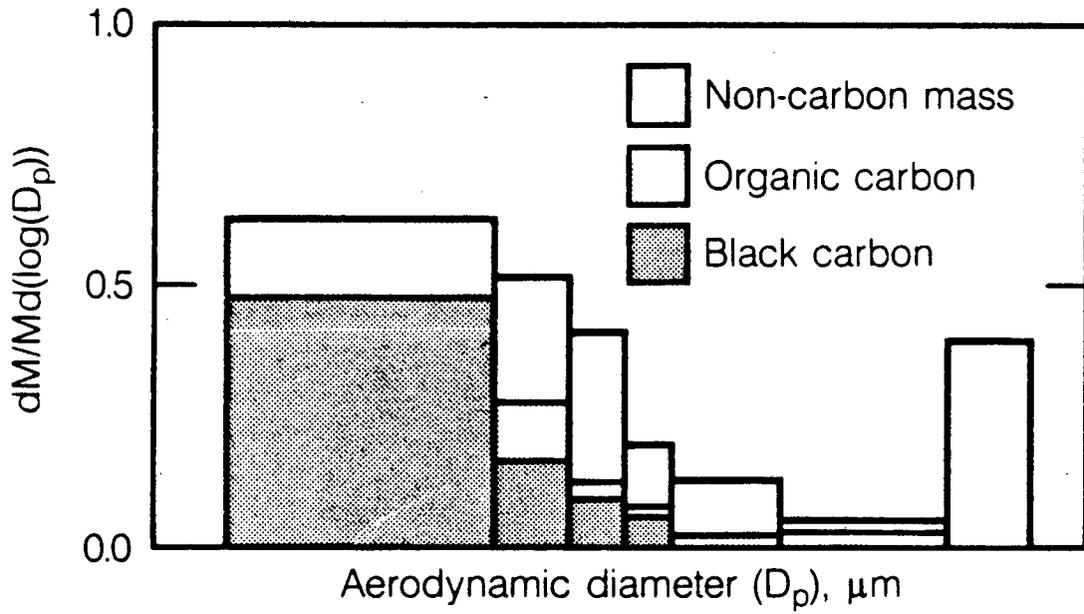


Figure 4

XBL 883-6021

Smoke particle distribution

3 wood cribs in room

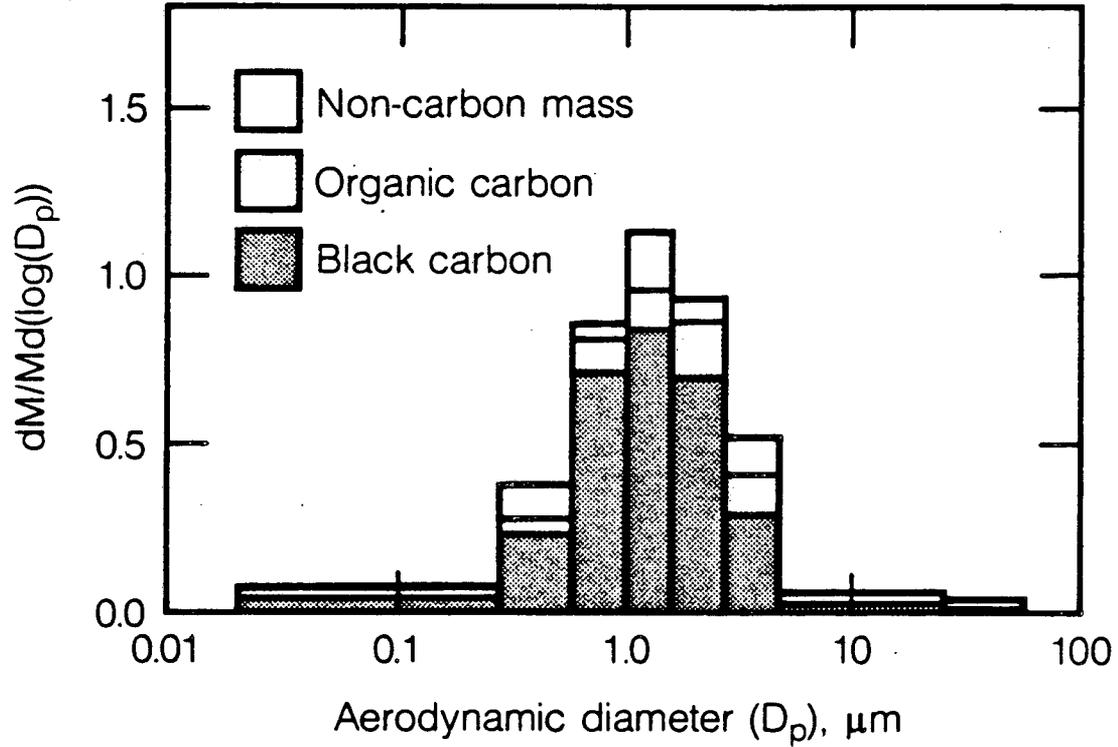


Figure 5

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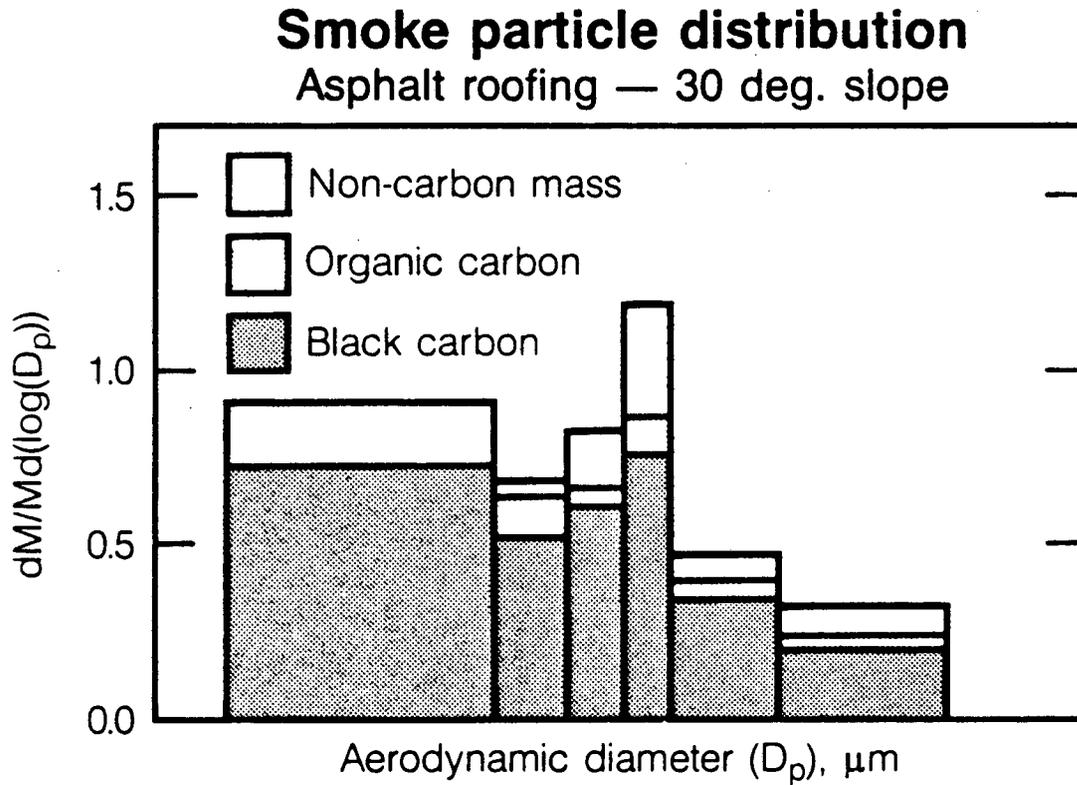


Figure 6

XBL 883-6018

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