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Smoke Particle Size Distribution in Pine Wood Fires

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ABSTRACT

There is a growing concern about the impact of ultra- fine particulates released from fires on the health of humans in fires and the related environmental pollution. However, there is no requirement to measure particle mass or number from legislated test fires and hence there is minimum information in the literature on this toxic hazard in fires. This work compares particulates generated from freely ventilated and restricted ventilation pine wood fires using the cone calorimeter. The standard cone calorimeter with freely ventilated combustion was modified by adding a discharge pipe to the cone heater that enabled direct fire product sampling from the cone outlet. The controlled atmosphere cone calorimeter was used for the restricted ventilation fire with metered air fed to the enclosure around the test area. Both tests used a radiant heat flux of 35kW/m². Real-time particulate number and size distribution were measured using the Cambustion DMS 500 particle electrical mobility spectrometer. The particulate size distribution showed a peak of ultra-fine aerosol particles of <100 nm in the early stage of the fire development and then changed to the larger size (100-1000 nm) with a peak of 200 nm as the fire progressed. The restricted ventilation fire generated more particles. There were high numbers of 20 nm particles throughout the fire and these have the greatest health risks. Toxic gases were also measured from the raw exhaust gases using a heated Gasmeter FTIR gas analyser.

KEYWORDS: Fire Particle size, Fire Toxic gases, Compartment Fires.

INTRODUCTION

Most fire deaths are due to smoke inhalation and associated toxic gases. Currently 'smoke' production is the only aspect of fire toxicity that is included in legislated fire tests of construction products, furniture materials, electrical cables, aircraft cabin materials and polymers. Smoke production in legislated testing is measured by optical obscuration and is related to visibility and impairment of escape through lack of line of sight to escape doors. The smoke regulations are not there because of the toxicity of smoke. The smoke production regulations have no requirement to measure the composition of the smoke either for soot particle size or toxic gases. The victims and some of the survivors of the Grenfell Tower fire in London had black lungs and some of those who survived had to have black particles flushed out of their lungs. The size of particles that reach the alveolar region of the lungs is <50 nm and particles of this size have no effect on light obscuration and so are not measured in the traditional fire smoke tests.

In the mid-1990s, epidemiological data in the USA and UK showed that 1% extra deaths occurred for every 10 µg/m³ of PM₁₀ in ambient air within days of the high particulates in the atmosphere [1-3]. Epidemiological studies have linked fine particulates in air pollution with cardiopulmonary mortality. The only medical explanation of this effect is that particles <50 nm must be present [4] as they cause alveolar inflammation and blood thickening [5], which reduces lung function and places a strain on the heart. This epidemiological data is based on correlations of PM₁₀ in the atmosphere and hospital admissions and asthma drug demand. The medical explanation [4] involves the

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presence of solid nano-particles in the alveolar region of the lungs and the composition of these particles is not significant [4]. However, other evidence of health effects of fine particles include allergic reactions, chronic obstruction pulmonary disease (COPD), pulmonary fibrosis and lung cancer [6], indicate that the chemicals absorbed on the particles may also be part of the health effects of ultra-fine particles.

There have been few studies of the ultra-fine particles generated in fires and they are not discussed in the fire literature as a cause of impairment of escape. However, in atmospheric particulate pollution the mass concentration limit is an annual 24 hour average of PM_{10} of $40 \mu\text{g}/\text{m}^3$. The present work will show that in wood fires huge numbers of particles are generated $<100 \text{ nm}$ and thus it is likely that breathing nano-particles in fires will be a major cause of impairment of escape and deaths in fires, that is currently not recognised. Gravimetric measurements [7] of particle mass in pine wood fires have shown that concentrations of particles $>0.1 \text{ g}/\text{m}^3$ occur in compartment fires, which is $>10^4$ times the atmospheric level of particle concentration where health hazards occur. In a fire, victims can breathe in one minute the equivalent of one day's exposure to contaminated ambient air. Thus it is clear that the breathing of fine particles in fires could be a significant cause of death and impairment of escape. Despite the health hazards and the associated impairment of escape due to fine particulates in fires, very little research has been done on fine particulate exposure in fires.

EXPERIMENTAL TECHNIQUES

Fuel properties

Wood is the most dominant combustible in fires, accounting for approximately 70% of CO_2 emissions and 65% of CO emissions [8]. Construction pine wood was used as fuel with characteristics shown in Table 1. Ultimate and proximate analysis were carried out using the Thermo Flash EA elemental analyser to determine the CHONS and the Shimadzu TGA-50 with a TA60WS processor to determine the water, volatiles, fixed carbon and ash content. The gross calorific value of the pine was determined using a Parr 6200 oxygen bomb calorimeter.

Table 1. Fuel properties

Parameter	Pine Wood Sticks
Proximate Analysis (Wt. %) (daf)	
Volatile Matter	86.54
Fixed Carbon	13.46
Ultimate Analysis (Wt. %) (daf)	
Carbon	53.95
Hydrogen	6.79
Nitrogen	0.11
Sulphur	0.00
Oxygen	39.15
Stoichiometric A/F by carbon balance	5.89
Gross Calorific value (MJ/kg)	18.9
Moisture (as received) (%)	6.18
Ash (as received) (%)	2.27

The cone calorimeter

The cone calorimeter (ISO 5660) is designed to measure the heat release rate and flammability properties of materials. The cone calorimeter reproduces the oxidative pyrolysis stage (class 1b) and well ventilated flaming fires (class 2) classification of ISO 19706 [9]. The authors have adapted the standard cone calorimeter [10] for direct raw gas analysis of the fire products at the outlet from the cone heater and used it with both free ventilation and restricted ventilation fires. The controlled atmosphere enclosure around the 100 mm square test fire in the cone calorimeter was used as a compartment fire with a metered air supply to the enclosure to generate a restricted ventilation fire, 3a classification of ISO 19706 [9]. It was operated with a controlled air supply designed to create rich combustion conditions that occur in air starved compartment fires.

The controlled atmosphere cone calorimeter with associated instrumentation is shown in Fig. 1. It was used for the restricted ventilation experiment to simulate an air starved compartment fire and for comparison with free ventilation, without the controlled atmosphere enclosure. The load cell, sample holder and cone heater were enclosed in the steel box, which had an air supply through the bottom of the air tight box. The airtight box was 38 cm long, 30 cm wide and 33 cm high, with a volume of 0.0376 m³. In the process of developing this experimental method it was observed that there was excessive heat loss from the metal box which influenced the temperature of the test specimen and hence influenced the heat release rate, HRR. The airtight box design was improved by insulating the inside and the door from the outside, using an insulation board to avoid heat loss. The combustion process was observed through a glass window on the cone calorimeter enclosure which was also insulated when not being used for viewing the flame. An insulation board was also placed in the sample holder underneath the test specimen to prevent heat losses to the supporting metal cylinder leading to the load cell heating.

The airflow introduced to the combustion chamber was from a compressed air supply and was measured using a variable air flow meter with the air flow varying from 6 – 28 L/min. This is 0.12 – 0.56 g/s air mass flow. The air mass flow per exposed surface area of the test specimen (0.01 m²) was 12 – 56 g/cm². The simple assumption that for all fuels there are 3.05 MJ of heat release per kg of air is another way of representing the ventilation conditions. This translates the air mass flow range into fire HRR in kW as 0.37 – 1.71 kW fires and in terms of the exposed surface area of the wood is 37 – 171 kW/m². In the present work a single air flow was used of 192 g/cm² or 58 kW/m² for the restricted ventilation condition. This is the maximum possible heat release rate (HRR) in the primary fire in the enclosed compartment.

Differential mobility spectrometer (DMS 500) particle size analyser

The Cambustion DMS 500 (Differential mobility spectrometer) was used to measure the particle size and concentration in real time. It measures particle sizes ranging from 5-1000 nm based on their electrical mobility. The DMS 500 responds to both liquid and solid aerosols. Liquid aerosols may occur from the volatile release from wood. Solid aerosols would comprise soot and ash particles. The DMS 500 is a transient instrument and can take numerous size distributions per second. In the present work, the size distribution as a function of the fire duration was measured.

Experimental procedure

The pine wood sample was exposed to the conical heater of the cone calorimeter radiating at 35 kW/m² for both free ventilation and with 192 g/cm² air mass flow in the controlled atmosphere cone calorimeter. 35 kW/m² is the standard heat flux for use in material evaluation for performance in fires using the standard cone calorimeter [10]. Also, 35 kW/m² was recommended by Flecknoe-Brown et al. [11]. Herzberg [12] noted that 35 kW/m² was a "trade-off between a lower value, which possibly would have caused materials to pyrolyse only, and a higher value which might have provoked an unrealistically clean burning behaviour".

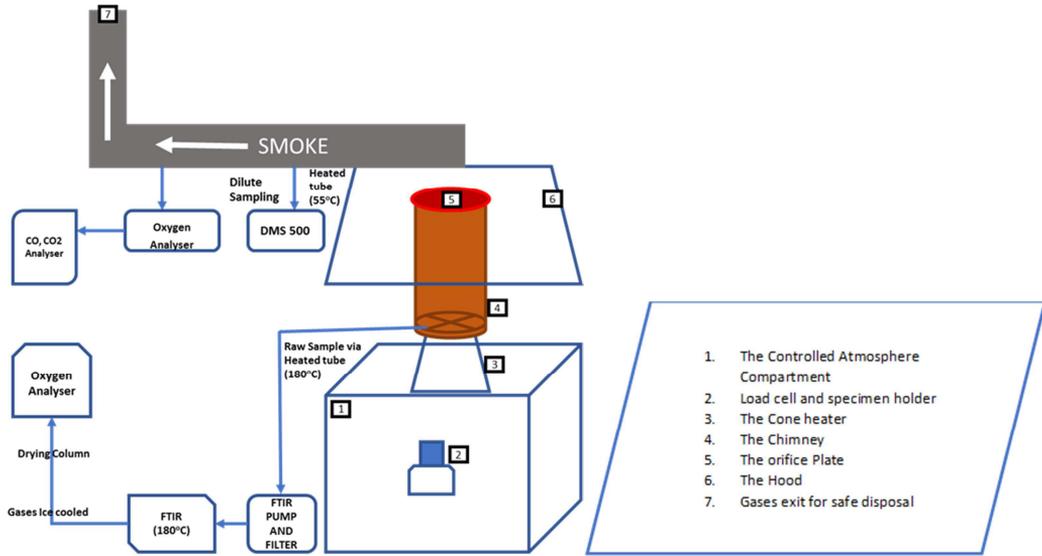


Fig. 1. The controlled atmosphere cone calorimeter.

Five rectangular shaped pine sticks (100 x 20 x 20 mm) were arranged horizontally in a 100 x 100 mm square sample holder of the cone calorimeter, exposing the top surface to the applied radiant heat flux. The initial weight of the wood was determined by the load cell as 127 g and 119 g for the two tests at different ventilations. The load cell was calibrated by checking with reference weights at the start of each test programme and was very stable. The 35 kW/m² radiant heat flux caused the thermal decomposition of the samples leading to auto ignition of the evolved gases and the auto ignition delay time was determined in the tests at 29 s for the restricted ventilation test and 192 s for the freely ventilated test. The fire continued until flaming combustion ceased and there was only char burning. It will be shown that this transition from flaming combustion to char smouldering combustion was associated with a change in the particle size distribution.

Sampling and analysis method

Both cone calorimeter experimental setups were modified to enable a raw gas sample to be obtained from the exit of the chamber. A 20 hole X sample probe was mounted on top of the exit plane of the cone heater for the sampling of the mean composition of the raw gases. A single hole probe had been used but was shown to not give a mixed mean sample. The X probe was mounted in a 76 mm diameter duct with flanges to join the cone heater exit to the chimney. This gas sample probe duct was 40 mm long. An 80 mm diameter chimney, the same diameter as the cone outlet, and 210 mm long was mounted on top of the sample probe and the total chimney length was 250 mm. It was found that oxygen was detected in the sample probe with rich mixture combustion, which should not occur. It was concluded that air was back flowing down the chimney, partially because the FTIR sample flow rate was similar to the air flow into the fire compartment. To avoid the air backflow, a grid plate restrictor was placed at the chimney exit to increase the exhaust flow pressure loss, which then prevented back flow of air down to the sample probe. This orifice plate on the chimney outlet had a 90% blockage of the flow and this prevented any oxygen occurring in the gas sample for rich combustion fires.

The schematic diagram showing the sample gas transport system to the gas analysers is shown in Fig. 2. The gases from the exhaust were transported via a heated 180 °C sample line to a heated pump (2.5-3 lpm) and filter. Another heated line was connected to the pump to transport the gases to the heated Gaset FTIR. The raw gas sampling was used because it prevented any post oxidation of

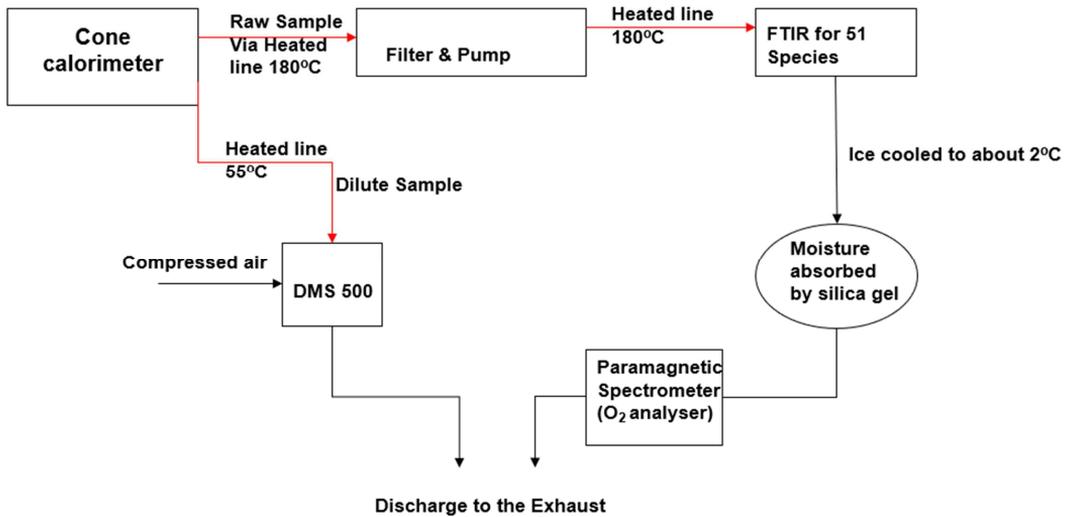


Fig. 2. Schematic of the analysers.

species by dilution of the gases with ambient air, as the gases were entrained into the cone calorimeter diluted flow metering section. It also prevented any losses of condensable gases that would occur if the exhaust was just thermally cooled [13]. The gas sample from the FTIR outlet was transported to a refrigeration cooler of about 2 °C via a PTFE tube and then to a silica gel column for the removal of water vapour before going into a paramagnetic oxygen analyser, after which it was discharged through the cone calorimeter discharge duct.

A dilute sample was used for the particle size analysis, as shown in Fig. 1. The standard dilution on the cone calorimeter was used and this prevents water vapour from the combustion products appearing as droplets, by cooling and reducing the concentration. Another reason for dilution is to reduce particle agglomeration, which affects the size distribution and reduces the number of particles. Kittelson [14] advocated that a dilution ratio of around 100/1 or more should be used. The products of combustion from the cone heater outlet (chimney outlet in the present case) are diluted with ambient laboratory air before entering the exhaust duct of the cone calorimeter. Thus, the gas sample for particle size analysis was taken from the cone calorimeter dilution tunnel.

A 55 °C heated sample line was connected to the dilution tunnel of the cone calorimeter to convey the sample to the Cambustion DMS 500 particle size analyser [15]. A heated line was used to allow dilution of the sample gas which serves to lower the dew point of the air so that condensation does not take place in the instrument. The DMS500 particle size analyser has an internal 10/1 diluter and the particle concentration provided by the instrument as output includes this dilution. The dilution from the chimney to the cone calorimeter was the ratio of the two air flow rates of 24 l/s for the cone calorimeter and between 6 and 28 L/min for the controlled atmosphere box or the air inflow for the standard freely ventilated test. This gave a dilution ratio range of between 240/1 and 51/1 for the controlled atmosphere setup. For the air flow used in the present work the cone calorimeter dilution was 146 for the restricted ventilation experiment and 26 for the freely ventilated experiment. The Cambustion number concentration was multiplied by 146 and 26 to give the concentration in the chimney. The overall dilution including the 10/1 in the DMS500 was 1460 and 260/1, which would ensure no size distribution change in the dilution system.

RESULTS AND DISCUSSION

Mass loss and heat release rates (HRR)

Compartment fires and freely ventilated fires are compared for the same pine wood test specimen at 35 kW/m^2 cone radiant heating and both had a gas sample for toxic gas and oxygen analysis taken as a raw heated mean gas sample from the chimney fitted on the cone exit. The results of the secondary combustion after air dilution beyond the chimney are not presented, apart from for the particle number analysis. Figure 3 shows that the ignition delay was much shorter for the restricted ventilation fire, due to the richer mixtures during the delay period, shown in Fig. 7, which have shorter ignition delays than the lean mixture for freely ventilated fires. Figure 3 shows the mass loss rate and total heat release rate for the primary combustion in the compartment. Both tests showed a mass loss rate at steady state of 0.07 g/s , with a much slower burn rate during the char burn phase from 1200 s. The two peaks in the mass burn rates will be shown to be associated with peaks in toxic gas emissions and in particulate emissions.

The oxygen mass consumption based heat release rate, shown in Fig. 3b, was computed from cone outlet chimney oxygen analysis, downstream of the FTIR. The wet based oxygen analysis (corrected for the water vapour removed based on the FTIR water analysis) is shown in Fig. 4. For the restricted ventilation fire Fig. 3b shows that the HRR peaked immediately after ignition and remained steady at 50 kW/m^2 , which was about one third of the freely ventilated fire where the steady state HRR was about 130 kW/m^2 . This was due to the low combustion efficiency with high CO , H_2 and HC emissions in the restricted ventilation fire due to the low fire temperature. Figure 4 shows that for free ventilation there was always surplus oxygen in the fire with high oxygen levels in the chimney. For the restricted ventilation fire the oxygen was close to zero for most of the time and combustion was completed in an external flame downstream of the chimney using the entrained oxygen from the dilution air.

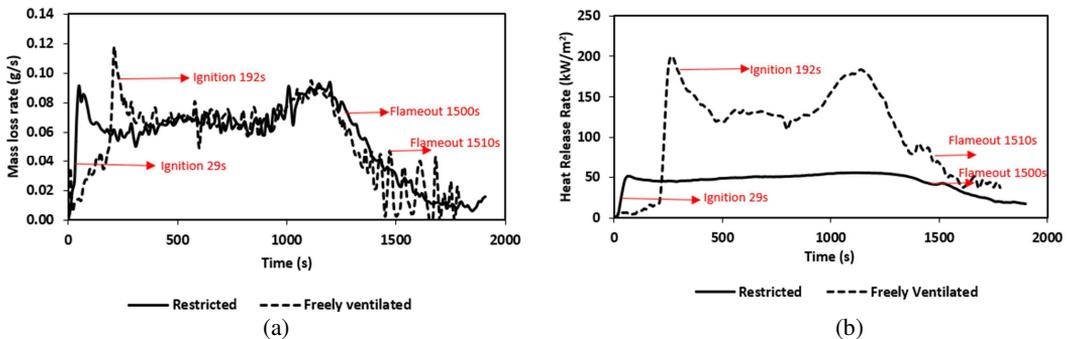


Fig. 3. Mass loss rate (a) and primary heat release rate (b).

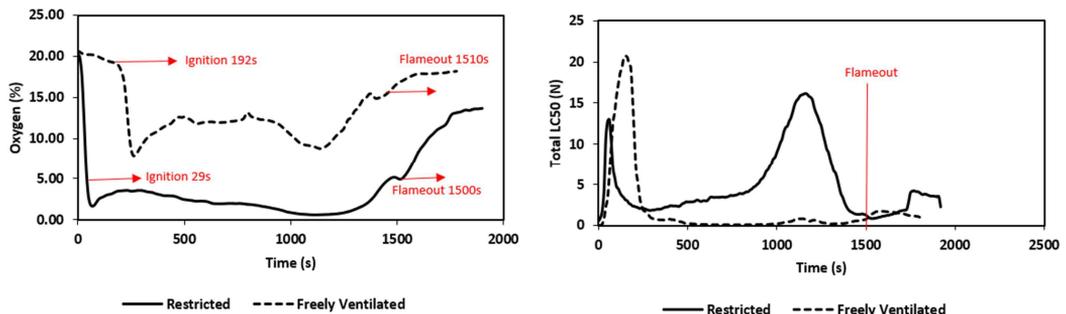


Fig. 4. Oxygen in the cone outlet chimney.

Fig. 5. Total LC_{50} FEC toxicity.

Toxicity, CO, hydrocarbon, fire equivalence ratio and combustion efficiency

All the FTIR measured toxic gases were divided by the LC_{50} limit and then summated to give the FEC total toxicity. This is shown as a function of time in Fig. 5, which shows both ventilations had a peak toxicity in the initial flaming combustion phase, but for restricted ventilation there was a second large toxic peak just before the flame out and a second smaller peak for free ventilation. These two peaks in toxicity occurred at the same time as the two peaks in the mass burn rate in Fig. 3 and the two minimum in oxygen in Fig. 4. The CO and total hydrocarbon (HC) yields are compared for the two fire ventilation conditions in Fig. 6 and they had a similar shape to the total toxicity results. The fire mean equivalence ratio by carbon balance and the combustion efficiency computed from the energy content of CO and HC are shown in Fig. 7. The restricted ventilation fire had rich combustion with an equivalence ratio of 2.0 throughout the flaming period. In the burning period after the ignition delay the CO and HC yields were much higher for the restricted ventilation fire as a result of the rich combustion. The freely ventilated fire burned lean with an equivalence ratio of 0.4 for most of the flaming period. The CO and HC yields were very low once combustion started after the ignition delay, as expected from the lean combustion. During the ignition delay period the CO yield was almost 3 times higher and HC was about 7 times higher in the freely ventilated test. This was due to the long ignition delay period of 192 s compared to 29 s of ignition delay for the restricted fire, where the products of wood decomposition were present but not ignited. The free ventilation dispersed the products of thermal decomposition, reduced their concentration and delayed their auto-ignition.

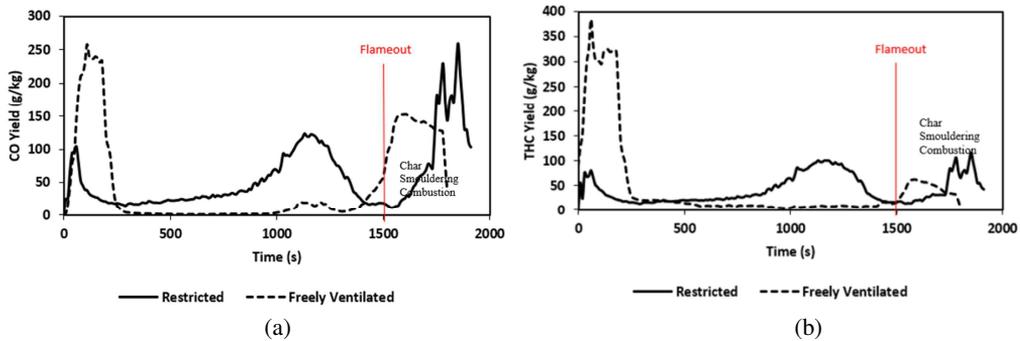


Fig. 6. CO yield (a) and HC yield (b).

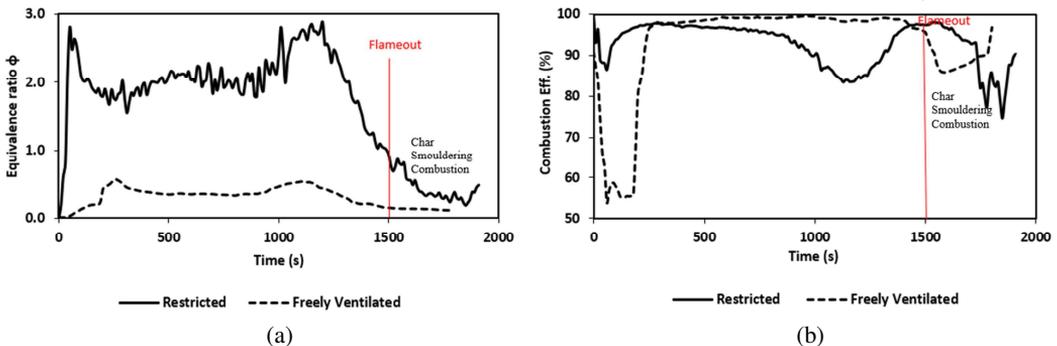


Fig.7. Equivalence ratio (a) and combustion efficiency (b).

Particle number concentration

The particle number concentration as a function of size and time is shown in Fig. 8. A bimodal distribution of the particle sizes was observed indicating the nucleation mode and accumulation

mode of the particle size distribution. The nuclei mode for both ventilation conditions was found to peak at 20 nm and the accumulation mode at 200 nm. The number of particles for the two modes, 20 nm and 200 nm are compared in Fig. 9 as a function of time for both ventilation conditions. The particle concentrations were highest at the peak HRR of 50 and 130 kW/m² for the restricted and free ventilated fires. For both ventilations, the 20 nm particle number was 10⁴ higher than those at the roadside [16], where the health hazards are known to be high. The effect of restricting the fire ventilation was to increase the ultra-fine particle number, due to the richer mixtures generated, as shown in Fig. 7. However, even for free ventilation with lean combustion the particle number was high at 1000 times roadside levels [16] for 20 nm particles. These results show that ultra-fine particulate emissions in fires from wood burning are a potential serious toxic impairment of escape hazard and materials should be evaluated for their propensity to form ultra-fine particles in fires.

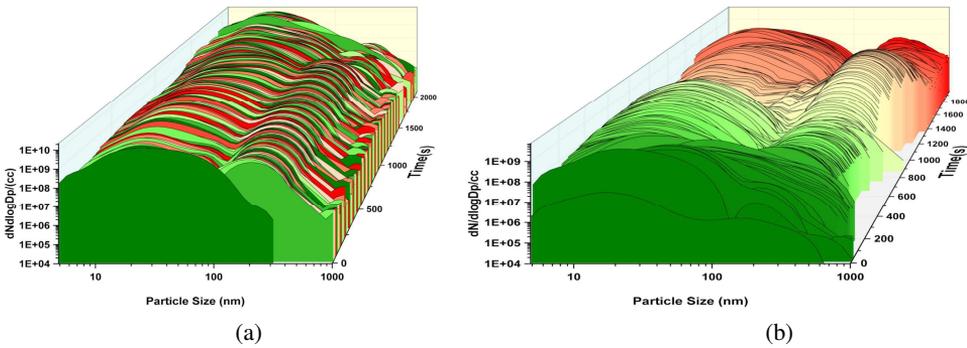


Fig. 8. Particle number concentration and size distribution; (a) restricted (b) freely ventilated.

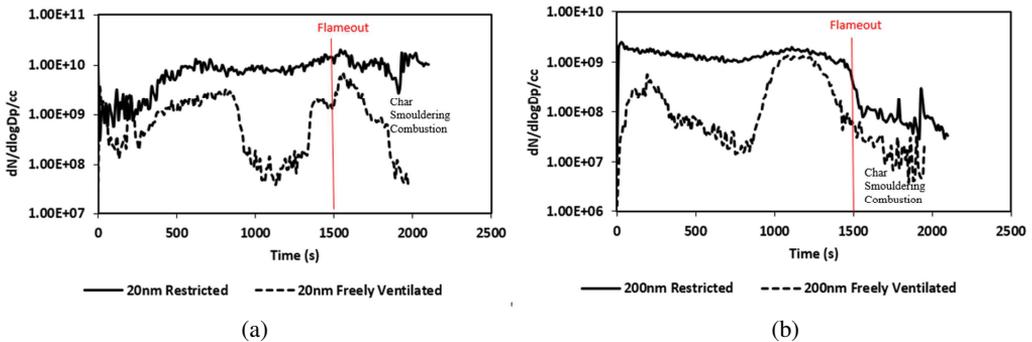


Fig. 9. 20 nm particle number concentration (a) and 200 nm particle number concentration (b).

Particle number comparison

The particle number size distributions as a function of time in the fire are shown in Fig. 10 for freely and restricted ventilation fires. Both fires showed a nuclei mode with a peak at 20 nm and an accumulation mode at 200 nm. These two size ranges are shown as a function of time in Fig. 9. This shows that for restricted ventilation the particle numbers were higher than for free ventilation. However, the time dependency was different with fairly consistent peak numbers at 20 and 200 nm throughout the restricted ventilation fire. For the free ventilation fire there were two peaks in the particle number of both 20 and 200 nm particles. These peaks in particle number coincide with the two peaks in HRR in Fig. 3b.

Relatively few investigations have been published on the particle size distribution in fires, especially in simulated compartment fires. Hertzberg and Blomqvist [12] used the low pressure

impactor particle size analyser, the Dekati ELPI, to measure the particle number from 60 nm to 10 µm size range for different fire materials, using the standard cone calorimeter. Figure 10 shows a comparison between the pine wood test at both ventilation conditions with the unspecified ‘wood’ measured by Hertzberg and Blomqvist [12]. All their particle size distributions were monomodal. At 200 nm, Hertzberg and Blomqvist [12] measured particle number of 10^6 p/cm³. In the present pine wood work, at 200 nm there were 10^9 p/cm³ for the restricted condition and $10^8 - 10^9$ p/cm³ for the free ventilation condition. These higher particle numbers for restricted ventilation were due to the richer mixtures where carbon formation occurs. The 5-60 nm size range of particles, which account for the greatest health risk, was not measured in Hertzberg’s work for ‘wood’ [12]. This size range of particles accumulates in the alveolar regions of the lungs, and such particles penetrate into the blood stream [1-6]. The ELPI particle size analyser does not have the size resolution, below 50nm where the greatest toxic particle hazard occurs and thus is not a good instrument for assessing particle size in fires. It is much better for larger particles >1µm of interest in optical obscuration.

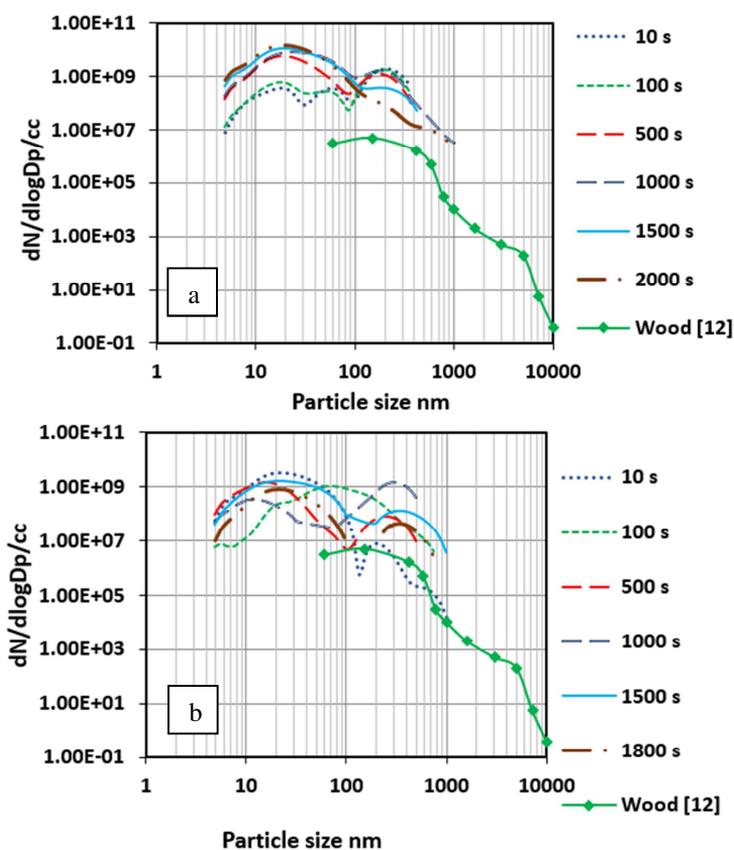


Fig. 10. Comparison of particle number distribution with that of Hertzberg [12] using the ELPI aerodynamic size separation instrument for restricted (a) and freely ventilated (b).

CONCLUSIONS

1. The particle size distribution of pine wood was measured in real time under restricted and free ventilation condition showing a bimodal distribution of nucleation mode and agglomeration mode.

2. The nucleation mode showed a peak of 20 nm on a number basis and a peak of 200 nm in the accumulation mode.
3. More particles were generated in the restricted ventilation fire due to richer combustion.
4. Concentration of ultra-fine particles generated in the pine wood test was much higher than that found in the literature. These ultra-fine particles could be a significant cause of death and impairment of escape in fires.
5. The cone calorimeter with ventilation controlled atmosphere around the combustion zone is good way to study the fire toxicity and particulate emissions of materials.

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