Size Distribution of Particles Emitted from Grass Fires in the Northern Territory, Australia

[1,2]*Arinto Y.P. Wardoyo,* [2]*Lidia Morawska*[*]*,* [2]*Zoran D. Ristovski,* [3]*Milan Jamriska,* [3]*Steve Carr and* [2]*Graham Johnson.*

[1] Brawijaya University, Malang, Indonesia
[2] International Laboratory for Air Quality and Health
Queensland University of Technology (ILAQH),
GPO Box 2434, Brisbane, Queensland 4001, Australia.
Phone: + 61 7 3864 2616 Fax : + 61 7 3864 9079 Email: l.morawska@qut.edu.au
[3] Defence Science and Technology Organisation (DSTO)

Abstract

This study investigated particle size distributions from the burning of several grass species, under controlled laboratory conditions, and also in the field, conducted during the dry season in the Northern Territory, Australia. The laboratory study simulated conditions such as burning phases and burning rate, and particle diameter differed depending on the burning conditions. Under fast burning conditions, smaller particles were produced with a diameter in the range of 30-60nm, whilst larger particles, with a diameter between 60-210nm, were produced during slow burning. The airborne field measurements of biomass particles found that under the boundary layer most of the early dry season (EDS) particles came from fresh smokes with a CMD of 83 ± 13nm, and most of the late dry season (LDS) particles came from aged smokes with a CMD of 127 ± 6nm. Vertical profiles of CMD showed that smaller particles were found higher within the atmosphere. These measurements provide insight into the scientific understanding of the properties of biomass burning particles in the Northern Territory, Australia.

Keywords: Biomass burning, particle size distribution, Northern Territory of Australia, airborne measurement, vertical profile.

1. Introduction

Among the many sources of air pollution, biomass burning has been identified as a major contributor of particles and gases in the atmosphere. Natural and anthropogenic types of biomass burning include forest fires, agricultural and waste burning, logging and land clearing slash, and burning for cooking and heating, Their emissions have played a significant role in influencing changes in atmospheric processes (Bodhaine 1983; Shaw 1987); causing the acidification of clouds, rain, and fog (Nichol 1997); and impacting on the transport of UV radiation within the atmosphere (Wurzler and Simmel 2005).

Knowledge of the characteristics of emitted particles has been identified as a very important element in developing a quantitative assessment of the impact of these fires. In addition to emission factors, which quantify the magnitude of emissions, understanding the size distribution of the emitted particles is also important, as it is size which determines particle dynamics in the air, as well as the impact on the environment. Previous studies showed that the majority of particles resulting from biomass burning were less than 2.5μm in diameter (Hays et al. 2002; Hedberg et al. 2002; Ferge et al. 2005; Wieser and Gaegauf 2005), depending on fuel variability, moisture content, burning conditions and burning processes. However, quantitative knowledge of their relationship with particle size distribution is still very limited. Several laboratory studies reported that particle size was proportional to flame size (Glasmann 1988) and fire intensity (Cofer III et al. 1996; Reid and Hobbs 1998), however the effect of the two together, in relation to particle size distribution, still requires further investigation.

Particle size distributions from the smoke produced by biomass burning have previously been reported in accumulation mode, with a count median diameter (CMD) in the range of 0.10-0.18μm. Smoke particles were reported to grow during transport in the

atmosphere and to increase in size with age (Reid et al. 1999). The growth of particles from biomass burning has been shown to occur within hours after emission (Abel et al. 2003) and also on a time scale of days (Radke et al. 1995; Reid et al. 1999). During the aging of the smoke, many species of secondary particles are formed, including those formed from organic acids (Gao et al. 2003; Haywood et al. 2003).

Characteristics of biomass burning particles have been studied in several regions of the world, showing that particle size varied between regions, however very few studies have investigated the characteristics of biomass particles in Australia. The first reported study was conducted over the Eastern part of the continent (Gras 1991) and later studies investigated smoke characteristics in the Northern Territory of Australia (Gras 1999; Tsutsumi 1999), focusing mainly on the measurement of scattering coefficients.

This paper presents the results of a study on particle size distributions produced by the burning of grasses in the savannah covering the Northern Territory, Australia. The experimental parts of the study included the controlled laboratory combustion of the grasses, as well as airborne measurements of the particle size distribution over of the Northern Territory, which experiences extensive fires every year. The aims of the study were to develop a better general understanding of the impact of fuel composition and burning conditions on particle size distribution, and also to provide more insight into the characteristics of particles emitted by biomass burning. This study was part of a larger Australian research project on particle emissions from biomass burning, involving the International Laboratory for Air Quality and Health, Queensland University of Technology (ILAQH), the Defence Science and Technology Organisation (DSTO) and the Australian Commonwealth Scientific and Industrial Research Organization (CSIRO).

2. Methods

2.1 Laboratory Measurements

The measurements of particle size distribution were conducted by burning grass samples collected from the Northern Territory savannah, under controlled laboratory conditions, designed to simulate the real life processes of fast and slow burning.

2.1.1 Experimental Setup

The experimental setup consisted of a burning system (modified stove), a dilution and sampling system, and a particle measurement system (Wardoyo et al. 2006). A modified commercial stove (66 x 74.5 x 55cm^3), fitted with a ventilation system to enable the introduction of a controlled amount of air into the stove, was used to simulate different burning rates. In order to obtain a homogeneous rate of air flow, the outlet of the ventilation system was connected to a rectangular hood, which was connected to a blower with a maximum capacity of 14 L/s, by a pipe 30 mm in diameter. The flow rate of the air was adjusted by a valve located at this connection.

The number concentration and size distribution of the particles produced during the burning process were measured using a Scanning Mobility Particle Sizer (SMPS) with a window of 10-600nm. The SMPS consisted of a 3071 TSI Electrostatic Classifier and a TSI 3010 Condensation Particle Counter (CPC). The sheath air flow was 4 L/s and the sample flow was 0.4 L/s. The scanning time and retrace time were 120s and 60s, respectively. Continuous measurements of the total particle number concentration were carried out using a CPC TSI 3022, with the sampling interval of 20s.

The smoke samples were taken from the flue through a probe 1cm in diameter and then introduced into an ejector dilutor (Dekati) where they were diluted 10 times with

heated, compressed, particle-free air to obtain a dry, diluted sample and prevent further coagulation. The sample was then mixed in a dilution tunnel with a constant flow of HEPA filtered ambient air get the concentration below 10^6 particles cm^{-3}. The flow rate and temperature of the samples were measured using an air velocity meter. The velocity of air in the dilution tunnel was kept > 1m/s to obtain a good mixing of the sample. The tunnel temperature was between 28°C and 30°C. The dilution ratio was calculated as:

$$DR = \frac{C_f - C_b}{C_d - C_b} \quad (1)$$

where C_f, C_d, and C_b are CO_2 concentrations measured using a flue gas analyzer and a TSI 8554 Q Trak Plus, in the flue, dilution tunnel and background, respectively (Wardoyo et al. 2006). Both instruments were calibrated prior to obtaining the measurements. The dilution ratio was found to vary from 100 to 200 depending on the burning conditions.

2.1.2 Sample Material and Preparation

Three species of grasses were selected from the savannah of the Jabiru area in the Northern Territory, in August 2005, according to their prevalence in the area (Wilson et al. 1990; Williams et al. 1999), as well as litter, containing a mixture of grass, leaves and branches. The grass species sampled were *Shorgum intrans, Aristida holothera* and *Eulalia mackinlayi*. The moisture content for each species was measured according to the difference between the sample weight before and after drying. The moisture content of the samples varied from 6-9% of wet mass for *Aristida holothera*, 7-10% for *Eulalia mackinlayi*, 6-11% for *Shorgum intrans*, and 8-15% for litter, and were similar to those reported for grasses growing in the savannahs of Northern Australia during the early and the late dry season, being 19% and 11%, respectively (Rossiter et al. 2003).

2.1.3 Burning Conditions

The samples were burned in the stove under 'fast burning' and 'slow burning' conditions. During fast burning, the stove was connected to a blower that introduced fresh air with maximum velocity 14 L/s. Under slow burning conditions, the blower was not connected to the ventilation system during the burning process. Burning of the samples was repeated three times, for each of the burning conditions.

2.2 Airborne Measurements

2.2.1 Study area

The large tropical savannah regions in the Northern Territory have observable wet and dry seasons. The dry season, which is mild to warm, occurs from May to October, and this is when annual uncontrolled fires usually occur (Gill et al. 2000). Mild intensity fires usually occur in the EDS, and high intensity fires in the LDS (Williams et al. 1998), with peak occurrences during July and September, respectively (Russell-Smith et al. 1997). The hot and humid wet season occurs from November to April, with an average annual precipitation of over 1000 mm. However, as a result of the Southern oscillation, the annual pattern of rainfall varies greatly from year to year (McKeon et al. 1990).

2.2.2 Measurement time and location

The airborne measurements of the size distribution of biomass burning particles in the Jabiru area were conducted during two campaigns in June and September 2003, which were in the early (EDS) and the late dry season (LDS), respectively. The measurements were performed during four 20 to 30 minute flights, either in the morning or in the afternoon. In June, the conditions were mostly fine and clear, while in September, they were mostly fine, with occasional cloud cover.

The preference was for afternoon flights, due to the increased number and intensity of fires that occurred during the day, however cloud cover conditions essentially determined what altitudes were flown, so this was not always possible. The altitudes flown were different between flights, as some flights were based on the collection of boundary layer data alone, whilst others were based on a combination of boundary layer and free troposphere data. The minimum altitude was set at 0.5 km, primarily for aircraft safety reasons, and the maximum altitude flown was 6.5 km.

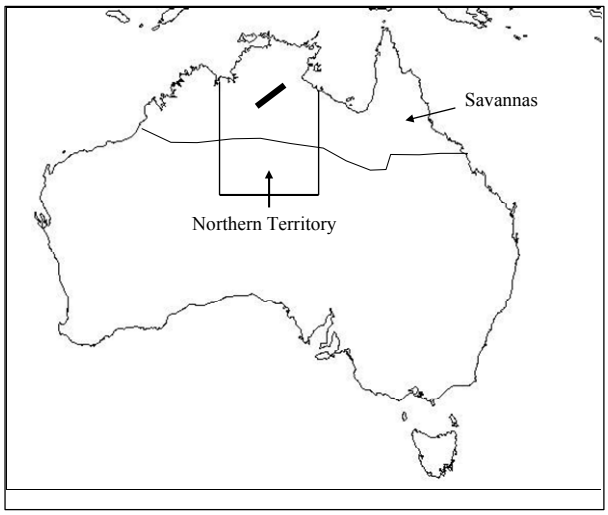

Figure 1. Savannahs in the Northern Territory (black line indicates flight path).

The airborne measurements were carried out along a vertical stack of horizontal flight legs, with the horizontal path originating from a point South West (SW) of 'Jabiru' (13.08 ^{o}S 132.32 ^{o}E) to a point North East of 'Jabiru' (12.11 ^{o}S 133.15 ^{o}E), in Kakadu National Park, Northern Territory (see Figure 1). The orientation of the flight path was chosen so that it was perpendicular to prevailing wind direction on the ground. The wind direction was predominantly from the South East (SE) in the morning, and although the wind direction varied from morning to afternoon, it only influenced the orientation of the flight path slightly. The majority of the fires were located on or near the flight path, with satellite data showing 39, 28, 72 and 41 hotspots detected on the 23rd, 24th, 26th and 27th

of June 2003, and 3, 11, 11, and 6 hotspots on the 22nd, 23rd, 25th and 26th of September 2003, respectively (http://www.sentinel.csiro.com.au). Although there were fewer fires in September, they were significantly more intense than those observed in June.

2.2.3 Data Analysis

For the laboratory experiment, the CMD of particles measured during the flaming and smoldering phases was statistically analyzed for each sample, to obtain the average CMD and standard deviation. For the airborne experiment, the average size distribution for each height region (defined in 3.2.2) was obtained by averaging the size distributions measured at several heights within the region, for each measurement day.

3. Results

3.1 Laboratory Measurements

3.1.1 Particle Size Distributions

Figure 2 presents the average CMD and standard deviation for particles emitted during burning. For the flaming and smoldering phases of fast burning, the CMD of the particles was 55 ± 7nm and 46 ± 3nm for *Aristida*, 36 ± 8nm and 50 ± 4nm of *Eulalia*, 50 ± 14nm and 37 ± 3nm for *Intrans*, and 37 ± 5nm and 77 ± 3nm for litter, respectively. For the flaming and smoldering phases of slow burning, the CMD of the particles was 122 ± 33nm and 60 ± 25nm for *Aristida*, 161 ± 20nm and 87 ± 13nm for *Eulalia*, 165 ± 50nm and 103 ± 25nm for *Intrans*, and 211 ± 44nm and 158 ± 29nm for litter, respectively.

It can be seen from Figure 2 that the CMDs emitted during fast burning show two distinct trends. *Aristida* and *Intrans* emitted larger particles during flaming and smaller particles during the smoldering phase, while it was the opposite for *Eulalia* and litter. It was also found that particle size distribution was unimodal for the flaming and

smoldering phases of fast and slow burning. Figure 3 presents an example of the average particle size distribution for slow burning.

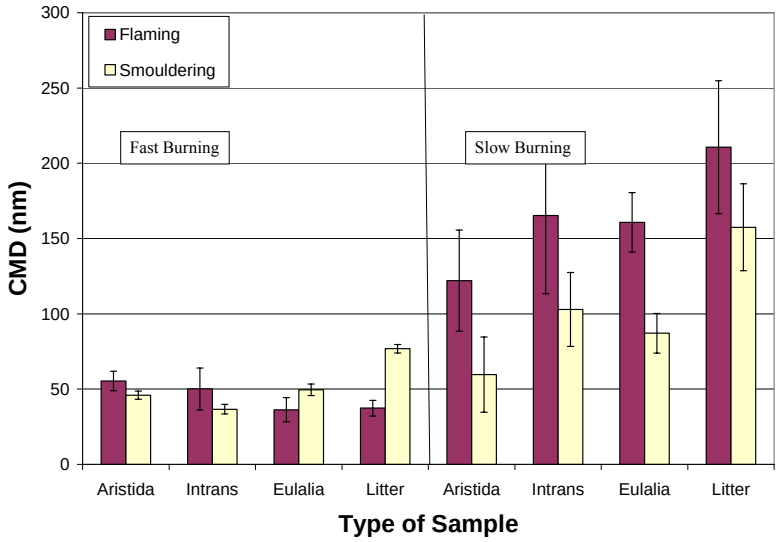

Figure 2. Count median diameter (CMD) of samples burned.

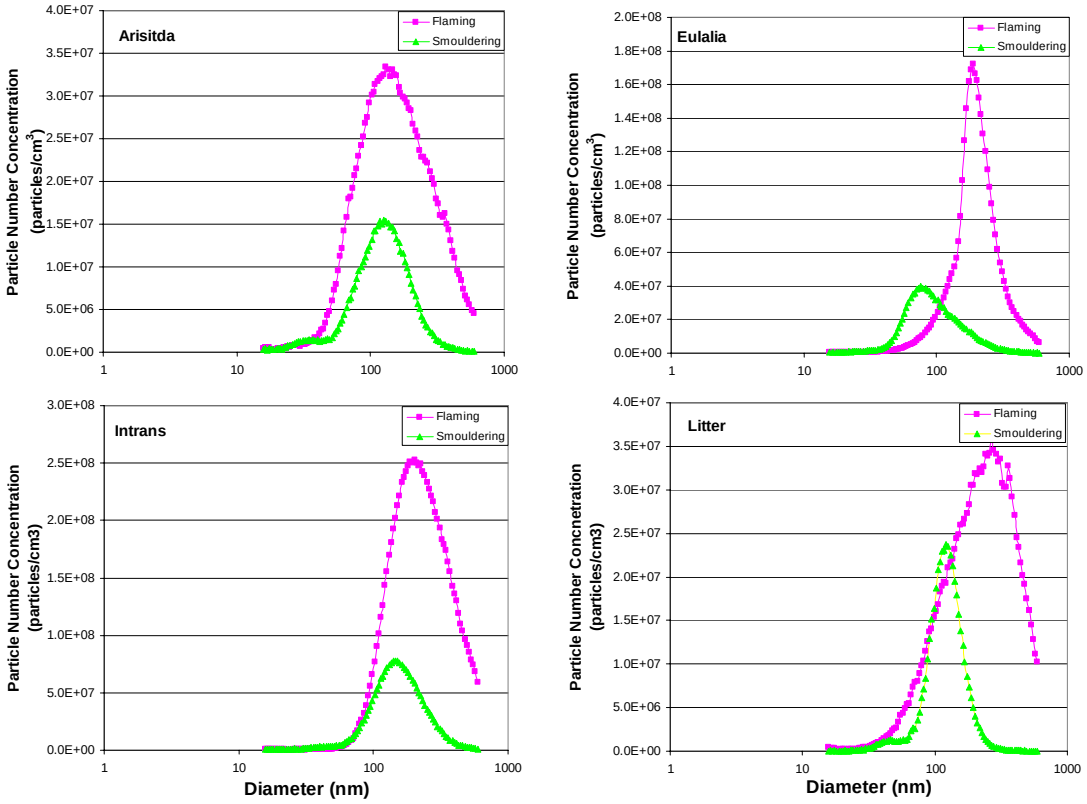

Figure 3. The average of size distribution for slow burning of grass samples.

3.2 Airborne Measurements

3.2.1 Boundary Layer Measurements

Vertical temperature profiles were used to estimate boundary layer height, indicated by a temperature inversion, or stable air, that demonstrates an increase in air temperature with an increase in height. In stable air, pollutants are trapped, which prevents them from dispersing into the free troposphere.

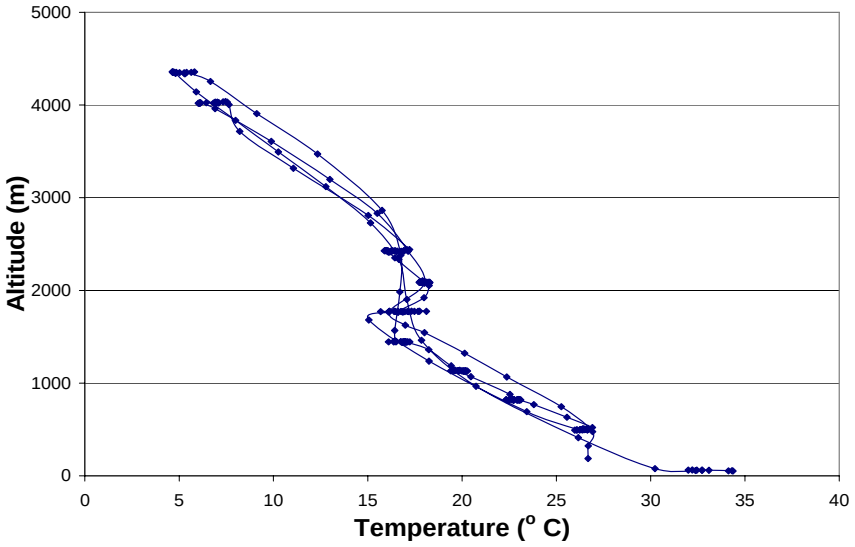

Figure 4. The temperature vertical profile measured on the 26th of June 2003.

Figure 4 is an example of the temperature vertical profile measured on 26th of June. The boundary layer was approximately 1700, 2000, 1800 and 1700 m on the 23rd, 24th, 26th and 27th of June 2003, respectively; 1800 m on 22nd of September; and 2000 m on 23rd, 25th and 26th of September 2003.

3.2.2 Particle Size Distributions

In order to characterize the particle size distribution obtained during the campaigns, the measurements were classified into three height regions, being region I - the lower

boundary layer (LB, calculated to be about 1700 m for the June campaign and 1900 m for the September campaign), region II - the upper boundary layer (UB, heights between 2000 and 3900 m), and region III - free troposphere (FT, height greater than 3900 m).

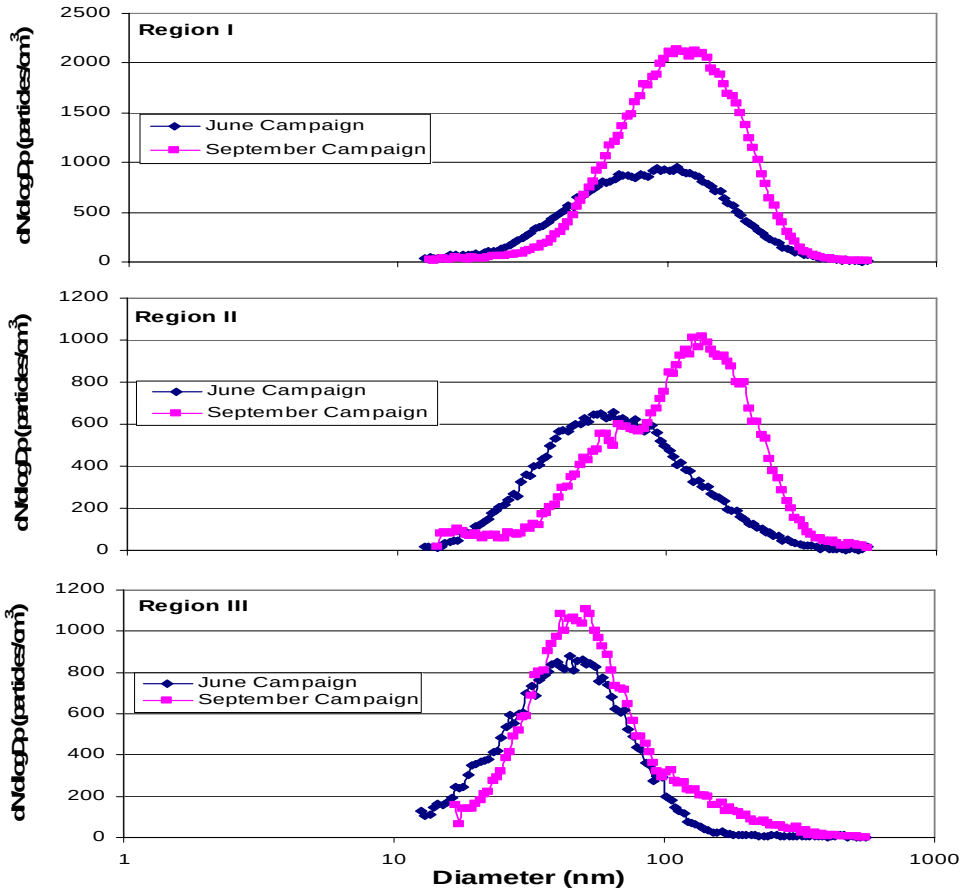

Figure 5. Average size distributions for June and September campaigns

Figure 5 shows the average size distribution of particles measured during the June and September campaigns for the three regions. For the June campaign (EDS), the peak particle size distribution is relatively broad for region I and II, with a significantly larger concentration of particles in region I. The size distribution of particles was significantly influenced by fresh smoke plumes from nearby fires, as indicated by satellite data which show many fires located close to the flight path (http://www.sentinel.csiro.com.au). During the September campaign (LDS), the peak particle size distributions are narrow,

indicating that the particles were more homogeneous in diameter across all three regions. This implies that the particles measured during the LDS were present in the atmosphere for some time prior to the measurements, which allowed for air mixing and other particle dynamics processes to occur. The satellite data showed that the only high intensity fires were hundreds of kilometers from the flight path (http://www.sentinel.csiro.com.au), thus the particles had likely originated from fires that occurred before the day of measurement.

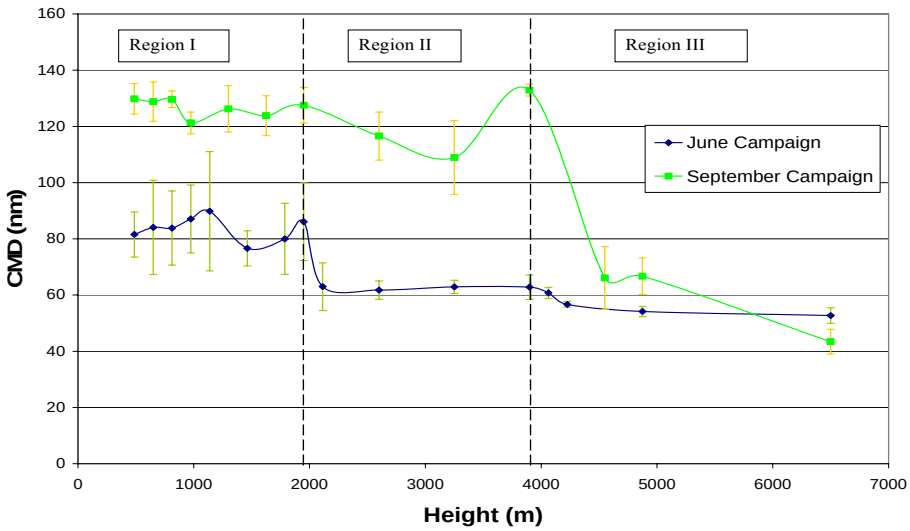

Figure 6. The measured CMD of particles during June and September campaigns.

Figure 6 shows the average and standard deviation of CMD for particles measured at different heights during the June and September campaigns. For the June campaign, the CMD in region I was larger than in regions II and III, with a relatively large standard deviation, as a result of the number of fires occurring in close proximity to the flight path. During the September campaign, the CMD for regions I and II were similar, but they decreased significantly in region III. The standard deviation of the CMD was relatively small, indicating that the sampled air was a well mixed, aged aerosol.

For the June campaign, the CMD in regions I, II and III were 83 ± 13nm, 68 ± 10nm, and 56 ± 2nm, respectively. For the September campaign, the CMD in regions I, II and

III were 127 ± 6nm, 119 ± 10nm, and 59 ± 7nm, respectively. Further analysis found that the CMD for the June campaign decreased by 18% from region I to region II, and also from region II to region III. For the September campaign, the CMD decreased by 6% from region I to region II and by 50% from region II to region III.

4. Discussion and Conclusions

4.1 Particle Diameter

This study investigated the particle size distribution originating from the burning of several grass species, under controlled laboratory conditions, and also during field campaigns, conducted during the Northern Territory dry season. Under real field conditions, biomass burning occurs mainly in the flaming and smoldering phases, with most emissions produced during these two phases. Therefore laboratory measurements were conducted during these two phases. Biomass burning also occurs under a variety of wind speeds, which is associated with variation in burning rate. As such, the laboratory study was also set up to include different burning rates, namely fast and slow burning.

4.1.1 Laboratory Studies

Results from the laboratory study showed that burning rate has an important impact on particle emission characteristics. Fast burning released smaller particles during the burning process, with an average CMD of 50nm, whilst slow burning resulted in larger particles, with a CMD of 165nm and 100nm, for flaming and smoldering, respectively.

This phenomenon was also reported in a recent study by Wardoyo et al. (2006) which investigated the size distribution of particles released during the different phases of burning. The study reported that the diameter of particles differed according to the speed of burning, with particle diameter ranging from 35-50nm for fast burning and 35-150nm

for slow burning. It was also found that particle diameter differed according to burning phase, ranging from 40-150nm for flaming and 35 – 50nm for smoldering, respectively.

A study conducted by Hueglin et al. (1997), found that the diameter of particles emitted from the burning of birch wood also differed according to the different phases of burning, producing particles with an average diameter of 170 and 60nm during the flaming and smoldering phases, respectively. The study also found that the size distribution of particle emissions was affected by air supply, whereby an increased air supply produced relatively smaller particles. A similar study by Hedberg et al. (2002), also investigating the particles emitted during the burning of birch wood, reported that the particle size distribution and number concentration depended on burning rate and the phase of the burning process, with most particles found to be in the range of 30-130nm.

A study conducted by Wieser and Gaegauf (2005), used several combustion systems with varying burning rates to show that particle size distribution was dependent on the type of combustion system. The study also investigated the effect of excess air on particle formation, and reported that a reduced air supply resulted in fewer particles, that were larger in diameter. Hays et al. (2002) studied the characteristics of particle size distribution by simulating the open burning of fresh green foliage and litter, in an enclosure with constant air circulation. The study reported that the size distribution was dependant on the phase of the burning process, with particle diameters of 100-150nm for the flaming phase and 70-150nm for the smoldering phase, depending on the fuel used.

In summary, previous laboratory studies found significant variation in particle characteristics dependent on vegetation type and burning conditions, with smaller particles produced during fast burning and larger particles during slow burning. The size

distribution of particles depended on burning phase only for slow burning, with relatively larger particles emitted during flaming. The results of this study confirm these trends.

4.1.2 Field Studies

The results of the field study found that particle characteristics differed across different periods of time during the dry season, with smaller particles measured during the EDS, compared to the LDS. The study also investigated the vertical profile of particles, and found that there was a consistent decrease in diameter as height increased.

A number of studies measuring particle size distribution from biomass burning have been conducted, finding CMDs of 20 ± 3nm in African savannah (Anderson et al. 1996); 120 ± 25nm in South Africa (Le Canut et al. 1996); 190 ± 3nm in the temperate forests of North America (Hobbs et al. 1996); and 120-230nm for aged smokes in Brazil (Anderson et al. 1996; Reid et al. 1998). Two separate studies in Amazonia reported different ranges of particle diameter: 15-279nm (Guyon et al. 2005) and 51-144nm (Krejci et al. 2005).

In summary, it can be concluded that the particle size distribution varied between the different locations due to a variety of factors, including vegetation, moisture content and weather conditions. The CMD of particles measured during Northern Territory field campaigns, conducted as a part of this study, were comparable to the lower values reported in the previous studies, ranging from 56-127nm across both the EDS and LDS.

4.1.3 Comparison between CMD measured in the laboratory and in the field

The CMD of particles measured during field campaigns in region I ranged from 83-127nm, for the EDS and the LDS, respectively, whilst the laboratory measurements found the diameter of particles varied from 50-165nm, depending on the rate of burning. Despite the unavoidable differences between laboratory and field conditions, these two

ranges are comparable, however this can not be used to verify the results, as there were many different factors influencing the conditions in the laboratory and the field.

4.2 Particle Vertical Profile

The airborne measurements of particle size distribution and concentration during the two field campaigns also provided information on the vertical profile of particles in the atmosphere. It was found that the concentration of particles decreased by 52% from region I to II, for both campaigns, as a result of the limited penetration of particles through the boundary layer. The reduction in CMD between the two regions, that is 20% in the EDS and 6% in the LDS, shows a difference in particle characteristics according to height. In the EDS, the particles were smaller and CMD decreased with an increase in height, however there was no significant difference in CMD between region I and II in the LDS, indicating similar physical properties of particles in both regions. This means that the particles measured during the LDS were well mixed and indicated that the particles were suspended in the atmosphere for a relatively long time.

A comparison of particle concentration between regions II and III showed that there were no significant differences during the EDS, and a small but statistically significant decrease in concentration of 13% in the LDS. However, large differences in particle CMD from region II to III, with the decrease of 18% in the EDS and 51% in the LDS, indicate that the particles in these regions have different physical properties and that the particles in region III were mostly existing free troposphere particles.

4.3. Particle Age

During transport in the air, particle size distribution is affected by the simultaneous generation and coagulation of particles and smoke mixing with the ambient air (Snegirev et al. 2001). The distribution is given by the dynamic equation (Seinfeld 1986):

$$\frac{\partial n(v,t)}{\partial t} = \frac{1}{2}\int_0^v \Gamma(v', v-v')n(v-v',t)n(v',t)dv' - n(v,t)\int_0^\infty \Gamma(v',v)n(v',t)dv'$$

$$-\frac{n(v,t)}{\tau_d} + \dot{n}_0(v,t), \qquad (2)$$

where $n(v,t)$ is the particle size distribution, $\Gamma(v', v-v')$ is the coagulation coefficient, τ_d is the characteristic time scale of particle dilution by ambient air, $\dot{n}_0$ is the particle generation rate, and *t* is the residence time (or the age of particles) based on time of movement from the generation zone to the detection point. Dilution time and particle age depend on the conditions of turbulent mixing, flow intensity and geometry of the compartment (Snegirev et al. 2001). Provided that the coagulation coefficient is constant, the differential equation of the particle size distribution can be derived from equation (2)

$$\frac{\mathrm{d}N}{\mathrm{dt}} = -\frac{\Gamma N^2}{2} - \frac{N}{\tau_d} + \dot{N}_0, \qquad (3)$$

where N is the total particle concentration, and $\dot{N}_0$ is the total particle generation rate. Assuming that the rate of particle dilution and particle generation are equal, and coagulation is the only factor influencing particle size distribution during transport, the total particle concentration can be presented by the coagulation equation (Hinds 1982):

$$\mathrm{N(t)} = \frac{\mathrm{N_o}}{1 + \mathrm{N_o K\, t}} \qquad (4)$$

where N(t) is the total particle concentration, K is the coagulation coefficient, and N_o is the initial particle concentration.

Although there are many other factors that could influence the particle size distribution, such as dispersion of the plume, sedimentation of particles and variations in fire size and intensity (Radke et al. 1995), to simplify and enable this assessment, those factors were assumed to be constant. By using the particle concentration data presented in

Table 1, particle age and initial particle concentration were calculated using equation (4), with the coagulation coefficient corrected as function of the diameter (Hinds 1982). Particle age and initial particle concentration were found to be 1.2 ± 0.5 days and 975 ± 40 particles/cm^3 for the EDS, and 6.5 ± 0.5 days and 2995 ± 260 particles/cm^3 for the LDS. These results show that particles found during the EDS were approximately 5 days younger than those found in the LDS. This can be explained by the fact that particles in the EDS originated from fresh smoke coming from sources that were close to the flight paths during the measurements. The age of biomass burning particles reported in other studies was found to be 2 days (Radke et al. 1995) and 5 – 15 days (Reid et al. 1999).

Table 1. Particle concentration measured during the campaigns

Campaign	Day of Measurement	Particle concentration in region I (#/cm3)
JUNE	23-Jun-03	898 ± 298
	24-Jun-03	913 ± 1000
	26-Jun-03	581 ± 179
	27-Jun-03	506 ± 104
SEPTEMBER	22-Sep-03	1393 ± 231
	23-Sep-03	1255± 309
	25-Sep-03	823 ± 91
	26-Sep-03	1280 ± 180

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