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Transcontinental Aerosol Measurements from Adelaide to Darwin

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ABSTRACT

Aircraft measurements of atmospheric aerosol were made during a number of transcontinental flights across Australia from Adelaide to Darwin during June and September 2003. A range of aerosol measuring equipment was operated during these flights enabling data to be collected on the aerosol chemistry, physical size distribution and scattering coefficient. Additional information has been obtained from other sources on the continental weather patterns and global atmospheric aerosol loading for the period that covers these flights. Together this information presents a picture of the aerosol sources, microchemistry, microphysics and optical properties allowing for differences to be explored between Southern and Northern regions of Australia at different altitudes (spatial information) and between the months of June and September (temporal information). The conclusions are that the aerosol mass density is small in general particularly during the month of June. At lower altitudes smoke tends to be the dominant accumulation mode aerosol and sea salt a major constituent of the coarse mode. There is evidence of some influence of intercontinental aerosol during September but the level of importance is difficult to quantify.

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Executive Summary

Task LRR 01/205 Atmospheric Aerosol Research was focused on identifying gaps in DSTO's knowledge about regional atmospheric aerosol. Aerosol plays a major role in atmospheric processes including influencing the Earth's radiation budget and as a result has an impact on global warming. Due to their scattering of electromagnetic radiation from the ultraviolet (0.01 microns) through to the mid wave infrared (5 microns) wavelengths aerosols also impact on the performance of military and civilian electro-optic (EO) sensors. In particular imaging surveillance sensors with a hyper-spectral capability can be strongly affected by the presence of atmospheric aerosol particularly those operating in the visible wavelengths.

This is the third report on measurements that DSTO has undertaken over the last two years to characterise the regional atmospheric aerosol. The primary aim of this work is to characterise the aerosol around the top end of the Northern Territory during the dry season when biomass burning is prevalent. This was mostly achieved by fitting purpose built aerosol measuring equipment to a commercial research aircraft that met the appropriate specifications to perform the task. This aircraft was contracted from Airborne Research Australia (ARA) a company based in Adelaide that specialises in carrying scientific payloads. Equipment was installed and flight tests were conducted in Adelaide before the aircraft flew a ferrying flight to Darwin, which served as the base for operations for the aircraft measurements that were conducted over Jabiru. The aircraft was carrying a full payload and was required to refuel at Alice Springs. The results reported on here were obtained during the transcontinental flights between Adelaide and Darwin during June and September 2003. As the aircraft was required to be ferried to Darwin with equipment already installed onboard it was decided to take advantage of this fact to make these additional aerosol measurements. The aircraft's flight profile was modified suitably to allow measurements to be made at a number of altitudes between Adelaide and Alice Springs and Alice Springs and Darwin. This was done in such a way so as to minimize the impact on the aircraft flight time and fuel efficiency and therefore did not add additional costs whilst still allowing useful data to be collected on the atmospheric aerosol properties. In this context these additional measurements can be considered opportunistic and will improve DSTO's knowledge on the nature of the atmospheric aerosol across the continent from North to South without requiring any additional resources. Essentially they are a free set of data.

This knowledge will be used by DSTO to improve our understanding and accuracy of atmospheric aerosol models that are used in prediction studies on the performance of EO sensors. This will enable DSTO to provide more robust advice to the ADO on the ability of EO sensors to meet performance requirements in the tropical North of the country when biomass burning is a dominant feature of the landscape and as far South as Adelaide. We see from the results of these transcontinental measurements that the air mass is exceedingly clean and hence based on these few measurements the impact on EO imaging

sensors from atmospheric aerosol particularly South of Alice Springs is not likely to be significant in general. This improved knowledge can also be used to influence development of new systems and the procurement of extant systems to meet the operational requirements of the ADO.

Authors

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Before joining DSTO as a Research Scientist in early 1998, Dr Carr completed an Honours Degree in 1991 and a PhD in 1996 both in Theoretical Physics at The Flinders Univeristy of South Australia. Shortly after joining the Weapons Systems Division he became Task Manager of the Infra Red Search and Track (IRST) task for the Navy which he worked on from 1998 through to the end of 2001. During his time in WSD he was involved with work on anti-personnel land mine alternatives and land based IRST as part of ground based air defence studies. Since joining ISRD in 2001 as a Senior Research Scientist he has been Task Manager for LRR 01/205 Atmopsheric Aerosol Research. This work has invloved experimental measurements both ground based and on an aircraft done in collaboration with CSIRO and University. He has also gained additional knowledge on EO and IR systems beyond wide band systems such as IRST to include hyperspectral systems in particular in both visible and IR wave bands.

Contents

1. Introduction

This report is the first to communicate the results of aircraft measurements that were conducted in 2003 to measure atmospheric aerosol. The primary purpose of this aircraft measurements program was to characterise the aerosol at Jabiru and surrounding areas in Kakadu National Park in the Northern Territory of Australia. The results of this work will be reported on elsewhere [1]. This activity follows on from work that was undertaken during 2002 to measure the ground based scattering coefficients at Jabiru over a three-month period [2]. This series of reports are outputs from the task LRR 01/205 Atmospheric Aerosol Research. These outputs aim to deliver knowledge on the fundamental microphysics, chemistry and optical properties of atmospheric aerosol primarily in the top end of the Northern Territory during the dry season when biomass burning is prevalent.

In this report the results obtained on four transit flights between Adelaide and Darwin are presented. The research aircraft used to conduct the aerosol measurements was based in Adelaide but was ferried to Darwin, which served as the base for operations for the Jabiru measurements. The aerosol measuring equipment was fitted to the aircraft and flight tests were conducted in Adelaide before the transit flight to Darwin. The opportunity was taken to conduct measurements during these ferrying flights to enable some characterisation of the aerosol across the Australian continent from North to South and at different altitudes.

This was a collaborative activity between the Defence Science and Technology Organisation (DSTO), the Commonwealth Scientific and Industrial Research Organisation's (CSIRO) Division of Atmospheric Research (DAR) and the Queensland University of Technology (QUT). DSTO contracted the services of Airborne Research Australia (ARA) to provide a suitable research aircraft, a pilot and an aircraft engineer.

Not all measuring equipment was used during the transcontinental flights. Enough data has been collected to present a useful picture of the continental aerosol microphysics, optical properties and some on the chemistry. Four sizing instruments were operating during these flights QUT's Scanning Mobility Particle Sizer (SMPS) [3], DSTO's Aerodynamic Particle Sizer (APS) [4] and two external probes (the probes) provided by CSIRO, the Active Scattering Aerosol Spectrometer Probe (ASASP) and the Forward Scattering Spectrometer Probe (FSSP) [5]. DSTO also operated its own three wavelength integrating nephelometer [6] during these flights. On some flights a CSIRO filter unit [7] was operated to collect particles for chemical composition analysis. Two filters were used a lower altitude filter (below 2438 meters (8000 feet)) and an upper altitude filter.

The data as presented here is preliminary. The outstanding issues to be addressed are the effect of particle losses through the isokinetic inlet [8] for the APS and the nephelometer and the correction of the APS and the probes size distributions taking into account ambient aerosol mass density and refractive index (RI) respectively. Without detailed knowledge on the chemistry of the atmospheric aerosols, determination of these last two quantities will not be possible. We have some data on the flight chemistry but insufficient

to allow for this type of analysis. Despite these issues the data can be used as we expect the SMPS data to be correct and hence any difference observed between the SMPS and the other sizing instruments can be attributed to the above effects. Hence the size distributions from these measurements can be used to provide a quantitative picture as to the nature of the size distribution across the continent. Additional corrections to the nephelometer data will be required to account for the angular truncation effect due to the restricted internal geometry of the nephelometer and also the effect of relative humidity (RH) changes. Analysis of the Jabiru data [1] indicates that these corrections are likely to be about 15% for a worst case where the ambient RH is large (c.a. 70%).

The implications of these results on atmospheric models such as MODTRAN [9], which is often used to determine the effect the atmosphere has on the performance of electro-optic sensors is left until later. The priority in this regard will be to use the Jabiru measurements [1] where the pay off to the Australian Defence Organisation (ADO) is expected to be the most in improving our understanding of aerosol properties in the tropical North of the country during the dry season and hence of our understanding of the impact they have on electro-optic (EO) sensor performance.

1.1 Background Material on Atmospheric Aerosol

For the sake of preciseness we take the following definition of what an aerosol is verbatim from the Guide to Global Aerosol Models (GAM) report [10].

An "aerosol" is a suspension of finely divided matter (liquid or solid) dispersed in a gaseous medium such as air. The suspended particles are called "aerosol particles" or "aerosols".

This definition is simplistic. The atmospheric aerosol is in equilibrium with its surrounding and often it can be in a gaseous, soluble or solid state and undergoing reactions between these three states. This depends on the chemical composition of the aerosols and the atmosphere. In particular it will depend on the ambient RH. A more accurate picture of the microchemistry is obtained by viewing the atmospheric aerosol as part of a chemical thermodynamic system in equilibrium ([11] Chapter 9). We cannot easily determine the chemistry of the specific aerosol compounds in situ. This is because it is necessary to collect the aerosol particles onto filters and hence alter the ambient conditions under which the aerosol normally exists. Further chemical analysis techniques involve determining individual ionic species, crustal material (soil) or elemental carbon, which involve breaking down the filter samples. The result of this analysis is a determination of the basic elemental composition of the aerosol (not including the organic carbon fraction which can be estimated from the missing mass). This information can then be used as input to a chemical thermodynamic equilibrium aerosol model along with the ambient atmospheric conditions (temperature and RH) to determine the actual concentration of the specific chemical compounds that compose the atmospheric aerosol.

There are two alternative views of an aerosol mixture. One is to assume that each aerosol particle contains only one chemical compound and the collection of such particles is referred to as an externally mixed aerosol. The second is to assume that each and every aerosol particle (for a given size range) is a uniform mixture of all the chemical compounds that compose the aerosol. This type of aerosol is called internally mixed. In the real world there are a range of other mixtures that span these two extremes. For this report we assume that the aerosol for each size range (fine or coarse modes) is internally mixed and hence in theory we can define an average aerosol density and refractive index per mode. In practice however we find that the flight chemistry does not provide us with a sufficient mass of material to allow for this level of analysis and we can only comment on what each mode is likely to be composed of.

To specify an aerosol uniquely requires information on the fundamental microchemistry as discussed above and also information on the size of the individual aerosol particles and the number of particles per unit volume i.e. the aerosol number concentration. Collectively these are referred to as the aerosol microphysics. It is understood that aerosols in the atmosphere that are either produced naturally or have anthropogenic origins do not have a precise size nor are they randomly sized. Aerosol particles fall into one of three or four size categories called modes where each mode is a distribution of particle size defined in terms of an equivalent particle diameter (De). These four modes are referred to as the nucleation, Aitken, accumulation and coarse modes [12]. The modes often have a lognormal form. In this case the mathematical form of the number size distribution per mode is given by (for the natural logarithm in this case)

$$\frac{dN(D;N_i,CMD,GSD)}{d\ln D} = \frac{N_i}{\ln(GSD)\sqrt{2\pi}} \exp\left[\frac{-(\ln D - \ln(CMD))^2}{2(\ln(GSD))^2}\right]$$

where D is the particle diameter, CMD is the count median diameter, GSD is the geometric standard deviation, N_i is the total number of particles in mode i where i = nucleation, Aitken, accumulation or coarse. In the case of the lognormal form there are relationships that exist between the total number of particles per mode and the volume of each mode (V_i) and between the CMD and the volume median diameter (VMD). For lognormal size distributions the above form gives the volume distribution by just replacing N_i with V_i and CMD with VMD.

The volume distribution can be given in terms of the number distribution in its most general form using the following equation

$$\frac{dV(D)}{d\ln D} = V_c \frac{dN(D)}{d\ln D}$$

where $V_c = \frac{\pi}{6} D^3$ if we assume the particles are spheres. Here c stands for the coarse mode for example. The assumption that particles are spherical is only made so that a simple relationship can be derived between the number and volume distributions. In practice the shape of particles can be irregular. In many applications that involve interaction with aerosol particles over large horizontal or vertical distances (such as scattering of electromagnetic radiation) the non-spherical nature of particles becomes less important as their orientation is averaged over. For some sizing instruments their response depends on the shape of the particles and hence ideally knowledge about aerosol particle morphology would be useful. This last equation does not assume the distributions are lognormal and applies equally well to the size distribution data from the sizing instruments. This relationship is used to convert the APS number distribution into a volume distribution. The results are given in section 5.2.

The particles that compose these modes are formed through different production pathways. The nucleation mode particles are formed through secondary gas to particle conversion whilst the coarse mode generally consists of primary material that has been mechanical produced through such processes as ocean bubble burst (sea salt) or wind lofting dust or soil. Another key point is that atmospheric aerosol number concentration changes with altitude and location. We make the distinction between the boundary layer (BL) aerosol (first few km) and then the free troposphere (FT) aerosol up to the tropopause. Above this altitude is the stratospheric aerosol that is mainly composed of volcanic dust and for the atmospheric layers above the stratosphere the majority of aerosol material has meteoric origins. Within the BL the horizontal variation in aerosol number concentration will depend mainly on local sources and to a lesser degree long-range aerosol transport. Above the BL the aerosol tends to be more horizontally uniform in its characteristics as the upper atmospheric chemistry and global weather patterns dictate the nature of the aerosol, both its microchemistry and physics.

The vast majority of aerosol numbers occupy the fine mode (< 1 micro meters (μm)), which is collectively used to refer to the nucleation, Aitken, and accumulation modes. Because these particles are so small the total mass (or volume) can often be weighted in favour of the coarse mode (> 1 μ m), due to the De^3 dependence assuming the particles are spheres.

Additionally aerosols are responsible for the scattering and to a lesser degree the absorbing of electromagnetic radiation and hence we need to know about the optical properties of the aerosol.

In this report we comment on the likely aerosol sources encountered during the transcontinental flights, what the basic chemistry was for each mode, the microphysics as described above and the optical properties. In the latter case we measured the aerosol scattering coefficient at three visible wavelengths (blue, green and red), which enables us to learn more about the nature of the underlying aerosol. We also probe how the aerosol properties change with altitude and location across the continent. Bringing together all this information to present a picture of what the measurements tell us about the underlying

nature of the aerosol is the main outcome of this report. To our knowledge there is no other atmospheric aerosol data obtained from aircraft measurements that exists in the literature and that covers the entire continent North to South passing over the centre at Alice Springs.

2. Flight Profiles and Instrumentation

Transit flights were divided into four transit routes two outgoing and two returning. Flights originated in Adelaide (Adel) and terminated in Darwin (Dar) with a single refuelling stop in Alice Springs (AS) on the outgoing transit. There were two measurement campaigns conducted in the top end of the Northern Territory during the weeks of 21-29 June and 20-28 September 2003. As a result there were a total of four transits between Adelaide and Darwin and hence eight separate flights (Table 1). In this report we give altitudes in units of 1000 feet or kilofeet (kft). The choice of kft was made as the aircraft's altimeter was in feet.

Table 1: A summary of the transit flights made during June and September 2003. T1= Transit route 1 e.t.c, Adel=Adelaide, AS=Alice Springs and Dar=Darwin.

Transit route	Orig-Dest.	Dates (2003)	Altitudes (kft)	Flight Leg Times (mins) 5, 10 kft	Flight Leg Times (mins) 15, 20 kft
T1	Adel-AS	June 21, Sept 20	5, 10, 15 and 20	15	20
T2	AS-Dar	June 21, Sept 20	5, 10, 15 and 20	15	20
T3	Dar-AS	June 29, Sept 28	5, 10, 15 and 20	15	20
T4	AS-Adel	June 29, Sept 28	5, 10, 15 and 20	15	20

Table 1 summarizes the transit routes, dates, altitudes and flight leg times for both June and September. The basic flight plan was to complete stepped descents into the destination for the transit route being flown. For example on the Alice Springs to Darwin transit route the flight path flown was to depart Alice Springs airport and climb to a cruising altitude of 23-24 kft and then commence a stepped descent, with steps at 20 and 15 kft each for 20 mins and then 10 and 5 kft each for 15 mins before descending directly into Darwin. These are approximate altitudes based on the aircrafts altimeter. There are differences between this altitude and the actual altitude based on the Global Positioning System (GPS) unit. The difference can be a few hundred feet but is not important given the altitude separation between flight legs is 5 kft. The choice of a stepped descent profile was made to provide sufficient time for the SMPS to stabilize and for the collection of multiple samples at each altitude. The impact on the aircraft flight time and fuel efficiency was marginal. This was chosen as the best compromise between the requirements of aircraft performance and the requirements of the aerosol sampling equipment.

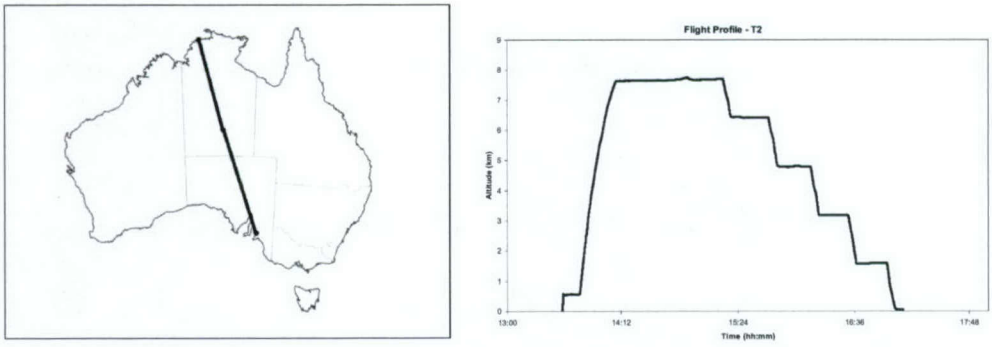

Figure 1: The complete transit route is overlaid on the map of Australia. The second figure is of a typical flight profile showing the stepped descent in this case into Darwin during June. This data comes from the CSIRO GPS unit fitted onboard the aircraft with altitude given in km.

The research aircraft was a Super King Air B200T operated under contract from ARA. This platform was ideal for these types of measurements as it had been fitted with aerosol measuring equipment in the past including wingtip probes and CSIRO's isokinetic inlet. More details about the aircraft configuration and the isokinetic inlet can be found in [1].

The APS and nephelometer were installed in a single 19-inch rack and mounted directly under the inlets that protrude into the cabin. These inlets are the sub sampling lines that reside inside the external shroud that together comprise the isokinetic inlet. Conductive silicon (consil) tubing was used to connect the inlets to the APS and nephelometer and to connect the outlets of the instruments to the exhaust manifold of the isokinetic inlet. The entire configuration provides a pressure seal. Besides being important for the safe operation of the aircraft it is obviously essential to prevent contamination of the samples with cabin air. The SMPS was also mounted in a single 19-inch rack. Consil tube was also used to 'plumb' the SMPS. The two CSIRO probes were mounted on opposite wing tips of the aircraft. The filters were mounted in a separate 19-inch rack and samples were drawn in through the isokinetic inlet and transported to the filter unit via copper and consil tube.

2.1 Instruments

All sizing instruments are particle-counting instruments. They record individual particle events (counts). Particle counts are then converted to a number concentration per bin size. The determination of the concentration of each sample for the SMPS and APS is achieved by maintaining a stable and constant volumetric flow rate given in terms of litres of air per minute. The ASASP and FSSP do not control the air flow as they sample directly from the atmosphere so in this case the sampling volume of each instrument needs to be well defined so that the total volume of air sampled can be determined. Raw particle counts are then converted to concentration and normalized to the logarithm (base ten) of the ratio of the bin lower and upper limits (diameters). The output of the APS can be given in multiple

formats [13] such as raw counts per bin, concentration per bin, cumulative concentration or the above-mentioned normalised concentration per bin. The latter is preferred so that once the diameters of the sizing instruments have been converted to physical diameter all distributions can be combined and compared irrespective of the bin spacing. This also allows the user to immediately see the underlying nature of the physical size distribution of the aerosol, which is typically lognormal or the sum of lognormals.

Thermo Systems Inc. (TSI) manufacture the SMPS, which is a configuration composed of a TSI 3010 Condensation Particle Counter (CPC) and a TSI 3080 Electrostatic Classifier (EC) with a short TSI 3081 Differential Mobility Analyser (DMA). Particles are sized as a result of the different electrical mobility particles of different size possess for a given electrical charge [3]. Briefly, the technique is sensitive to particle sizes from 0.01 μm up to 1.0 μm. Initially the particles are drawn into the electrostatic classifier where they are given a charge of +1 coulombs before being transported in the airflow to a region between two cylindrical metal plates that are charged at different potentials. The electric charges are produced according to a Boltzman distribution and hence there is a non-zero but small probability of multiple charges being deposited on a single particle (especially for large sizes). The instrument takes account of this effect by using a multiple charge correction algorithm. The charged particle is deflected toward the plate with the opposite charge (the inner plate) with the amount of deflection depending on the potential difference between the plates and the electrical mobility of the particle, which for a given charge is a function of the size of the particle. The classifier then scans the size range by varying the potential difference between the two plates causing particles of successive size to pass through a gap in the inner plate to be transported to the CPC. In the CPC the particle is coated with butanol as a vapour that 'adheres' to the outside of the particle increasing its size such that it can be registered by optical means. In combination the EC and the CPC are referred to as the SMPS.

This technique does not suffer from the problem of the need to convert the distribution to a physical diameter, as the electrical mobility is a function of the physical size of the particle. However because the entire size range must be electronically scanned this means the shortest sample time is 150 seconds of which 120 seconds involves the actual measurement of the size distribution. This limits the horizontal sample size on a moving platform and hence the number of samples that can be obtained. At low ambient pressures such as those experienced sampling from an aircraft at altitude the lower limit of the instruments sizing is restricted due to the maximum potential difference that the air at low pressure can 'support' before being electrically broken down and forming a dis-charge and hence an arc between the two plates. Therefore at the higher altitudes some smaller sizing capability is lost.

The SMPS directly provides the physical diameter size distribution and hence without the need for any corrections is the most accurate representation of the physical size of the aerosol being sampled. This is not the case for the APS or the two external probes.

The APS is a TSI model 3321 and it sizes aerosols by determining the time of flight of a particle in an accelerated airflow [4]. The response of the particle to the airflow is a function of particle size and shape and since the moving air (sonic velocities) is outside the Stoke's regime it also depends on the mass density of the particle. The majority of atmospheric aerosols have a density in the range $0.8 \leq \rho \leq 2$ g cm^{-3} where the effect of the non-stokesian nature of the airflow does not affect the sizing of the particle. The instrument measures aerodynamic diameter (D_a), which is an equivalent diameter, defined to be the diameter of a sphere with unit density that has the same aerodynamic properties as the actual particle with a given physical diameter (D). The equivalence in the aerodynamic behaviour of the two particles is often stated as the two particles have the same gravitational settling velocity. The advantage of measuring aerodynamic diameter is that the mass density of the particle is not needed a priori to accurately measure the size distribution (allowing for stokes correction as alluded too above). If we are able to determine the average aerosol mass density through chemical analysis or some other means then we can convert the aerodynamic diameter distribution to a physical diameter distribution during post processing of the data. To do this we make use of the following equation

$$D = \frac{1}{\sqrt{\rho}} D_a ; \ \rho \text{ - average aerosol density}$$

leaving out the dynamic shape factor and the Cunningham slip correction factor [14]. The Cunningham slip correction factor should be close to unity for particles larger than 1 μm. Shape effects may well be important for particles that are not spherical in appearance such as square shaped salt crystals. These form a significant fraction of the coarse mode aerosol.

The majority of sampling losses for the APS will come through the isokinetic inlet. This inlet has been tested and has been shown to sample representatively out to 3 μm. The APS is designed to measure from 0.5 to 20 μm (aerodynamic diameter) but realistically the number concentration above 10 μm in the atmosphere in general is very low. The region between 3 and 10 μm is where the most noticeable effect of sampling losses is evident. In this case we should look to the FSSP probe data to more accurately determine the coarse mode tail for 5 μm and above. However the lack of coarse mode aerosol to begin with means that this is a very challenging problem within the sensitivity limits of the instruments available.

Particle Measuring Systems (PMS) manufacture the ASASP-100X and FSSP-100, with newer models now produced by Particle Metrics Inc (PMI). The probes as the names suggest detect particles by optical means [5]. The ASASP detects particles in the size range 0.3 - 3 μm and the FSSP 2 - 47 μm. They are both forms of optical particle counters with the advantage of being externally mounted and hence require no sampling lines. The drawback is that they are measuring aerosols in a sample of air moving at large speeds and hence the response time is limited but the advantage is that the sample is measured at the ambient conditions (temperature and RH). The intensity of scattered light is a function

of the particle size, shape and refractive index. The internal processing of the instrument is based on the assumption that the particles are spherical in shape and hence that the scattered light intensity is given by Mie theory for scattering from a uniform dielectric sphere. In this case the complex refractive index and the diameter of the particle determine the amount of scattered and absorbed light for a uniformly mixed aerosol particle. A He-Ne laser operating at a wavelength of 6328 angstroms is used to illuminate the particles. The instrument is calibrated using spheres of a known size and refractive index. The problem with this approach is that the ambient aerosol refractive index is likely to be different from the calibration particles. This can be corrected for in post processing once an estimate for the mixed aerosol refractive index is made from any available chemistry data. No account is taken of non-spherically shaped particles.

The nephelometer is a TSI model 3563. It measures the atmospheric aerosol scattering coefficient at blue (450nm), green (550nm) and red (700nm) wavelengths [6]. It is a scattering chamber illuminated with an approximate cosine light source with a light trap at one end and photo-multiplier tubes (PMT) at the other. Particle laden air is drawn into the chamber via the inlet port and exhausted via the outlet. A blower that comes attached to the instrument generates and controls the airflow. More details about the nephelometer operation can be found in [2].

3. Environmental Conditions and Aerosol Loading

The purpose of this section is to highlight the most likely sources of aerosol that were encountered on the transit routes. In section 4 we present the flight chemistry data, which allows us to quantify what the basic chemical components are on a mode-by-mode basis. The small amount of mass that was collected precludes any more detailed analysis than this. For example we find that it would be difficult if not impossible to use this chemistry data to determine an effective aerosol density and refractive index for each mode. This does not mean however that our results are not useful. When combined with information about the size distribution and scattering coefficients we can learn a great deal about the properties of the transcontinental aerosol. With additional knowledge about the nature of the landscape we were flying over, prevailing weather patterns and other more apparently random events such as fires we can attempt to make a reasonable assessment as to the most likely composition of the aerosol. This is done on a mode-by-mode basis by assigning each aerosol type to one of the nucleation, Aitken, accumulation or coarse modes. The most obvious sources being biomass smoke, sea salt, soil and desert dust (or sand). There may be some impact from urban centres indirectly through long-range transport as well as from intercontinental smoke and dust.

The transit route from Adelaide to Darwin covers a range of rural landscapes from the developed and farmed regions North of Adelaide to the semi-arid and desert regions of Northern South Australia and the Northern Territory finally to the tropical savannahs encountered in the top end of the Northern Territory. The coarse mode aerosol is

composed primarily of particles that have been produced directly through mechanical means such as wind lofting soil and sand from the desert regions into the atmosphere. The wind varies with altitude both in speed and direction. It is the ground level wind that is responsible for the coarse mode (BL) aerosol generation and the higher altitude winds for the long-range transport of aerosol that were generated elsewhere on the continent or further a field.

The aerosol in the FT will for the most part have been generated through secondary formation mechanisms such as gas to particle conversion. The upper level chemistry will primarily determine the composition of the FT aerosol. This should be similar across the continent allowing for differences in the height of the inversion and some BL (ground level generated) aerosol extending through the inversion into the FT.

The weather conditions alter dramatically from South to North. June is the start of winter in Adelaide and the start of the intense part of the dry (biomass burning) season in the top end of the Northern Territory. Conditions across the continent change as well particularly wind direction at lower altitudes, which has an impact not only on coarse mode aerosol production at a specific location but also on the long range transport of the aerosols. At higher altitudes in the FT winds tend to be more uniform both in terms of speed and direction across the continent (Figure 2) and the fine mode aerosol can be transported long distances in the period of days and weeks over its lifetime. Local weather conditions have a major impact on measurements when there exist local aerosol sources such as biomass fires. In this case it is relatively easy to correlate what is observed with the sizing instruments and the nephelometer to specific fire activity. Higher in the troposphere aerosols may well have travelled from thousands of kilometres away. In this case the history of the air mass trajectory over the preceding days or weeks is important to trace the long range transport of the aerosol being sampled and possibly the origin or source of the particular aerosol. For example Saharan dust from North Africa has been transported as far as the Caribbean. Similarly biomass smoke from South America has been transported vast distances into and across the Southern Atlantic. We have not undertaken an air mass back trajectory analysis but instead rely on mean and anomalous weather conditions for the months of June and September (Figures 2 & 3) plus knowledge about the likely sources of aerosol.

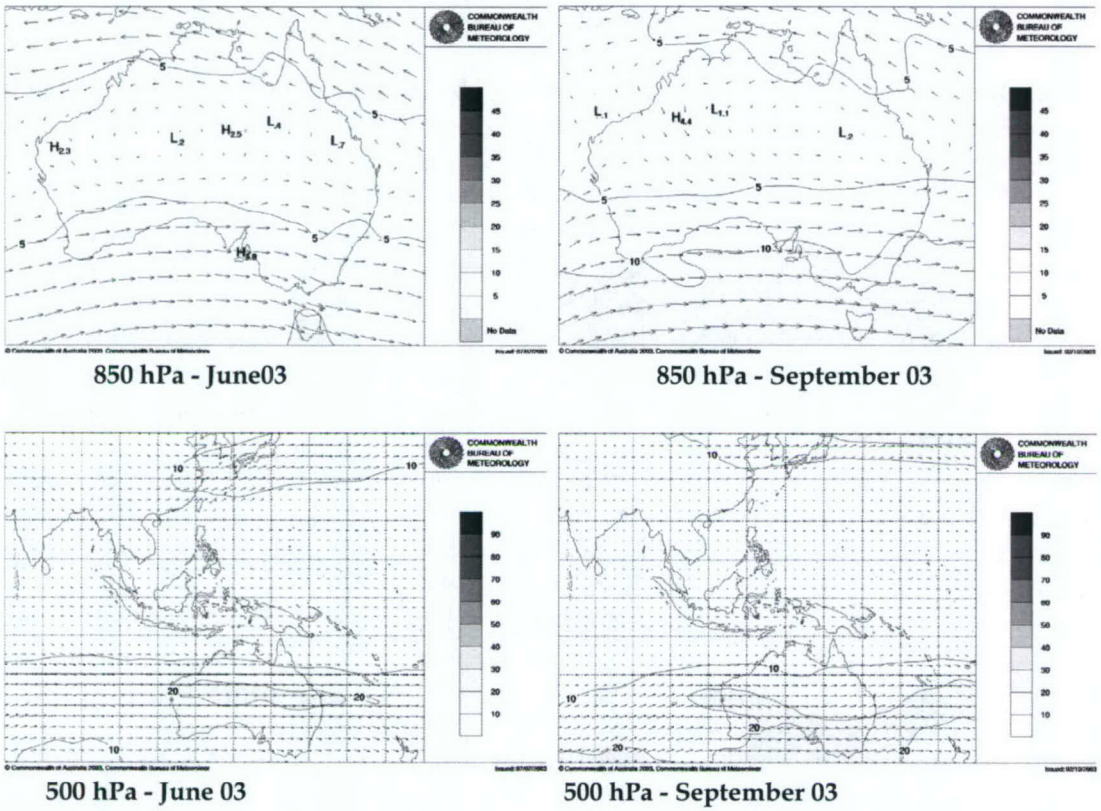

Figure 2: Monthly Mean Horizontal Wind patterns for Australia and the Asia Pacific region. At the higher altitude the horizontal wind becomes more uniform both in terms of strength and direction. The pressure is given in hector Pascals (hPa). These pressures correspond to approximate heights of 5kft and 20 kft respectively. (© Commonwealth of Australia 2003, Commonwealth Bureau of Meteorology)

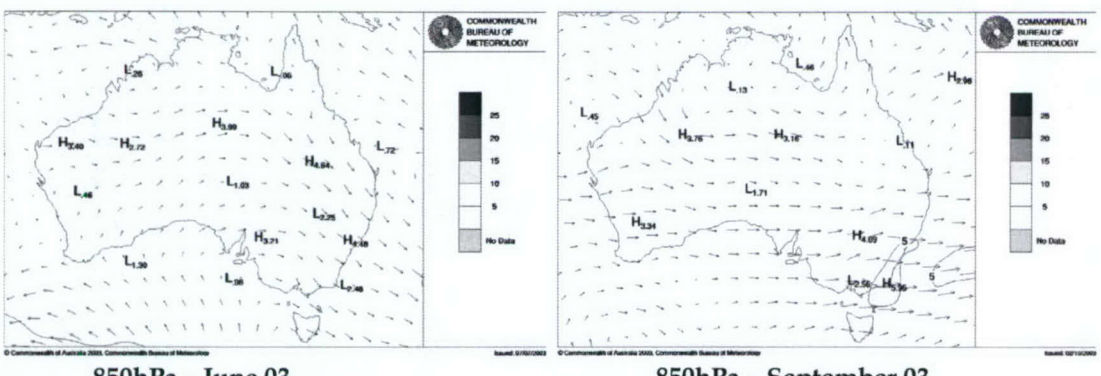

Figure 3: Monthly Anomaly Horizontal Wind patterns for Australia. (© Commonwealth of Australia 2003, Commonwealth Bureau of Meteorology)

The following information about these maps and the associated Global Analysis and Prediction (GASP) model are taken verbatim from the Bureau of Meteorology website.

These maps showing broad scale atmospheric circulation patterns are monthly mean and anomaly maps, showing analyses derived from the Bureau of Meteorology's GASP model. The 00Z analyses for each day in the month are used to generate the monthly fields. The monthly maps are typically generated in the first few days of the following month. The global model GASP uses spectral numerical techniques and is currently run with triangular wave number T_L239 truncation (approximately 85 km horizontal resolution). The model's domain is global, and has 29 levels from the surface to 10 hPa. Analyses of observations are made at 6-hourly intervals and twice per day forecasts are generated out to 7 days. The GASP system provides boundary conditions for the limited area systems and also provides the surface winds used in the global sea state prediction scheme.

Examining Figure 2 (reproduced here with permission of the Commonwealth Bureau of Meteorology [15]) from the two bottom maps we observe that the air mass for the higher altitude emanates from the West of the continent (with a small Southerly component most noticeable in September) and most likely is relatively clean air with possible Antarctic origins. There is also long-range transport of intercontinental smoke and dust from Africa for example (Figure 8). Perth is the only major urban centre located on the continent West of the flight paths. This explains the very low levels of aerosol present.

We can use fire maps (Figures 4 and 5) for the top end of the Northern Territory to allow us to locate on any given day the number and location of fires with respect to the flight path. Smoke is the dominant aerosol that is present in the Northern transit routes T2 and T3. A significant fraction of the coarse mode aerosol is likely to be sea salt and wind blown soil or dust and for some of the flight paths possibly desert sand.

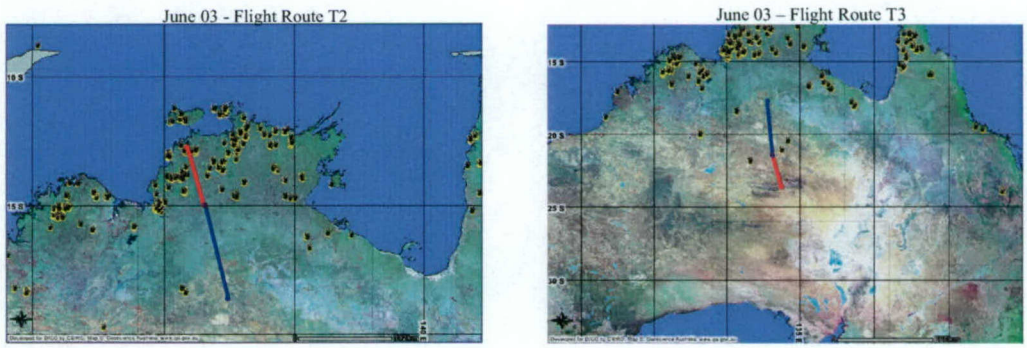

Figure 4: June 03 transit routes T2 and T3. The red line (shorter line segment) shows the location on the ground over which the 5 and 10 kft flight legs were flown. The blue line shows the same for the 15 and 20 kft legs. The cluster of fires located just below the red circle (termination point for transit route T2) is the source of the large accumulation mode counts for the 5 kft leg. The black and yellow markers indicate fire spots detected over the period 20-22 June 03.

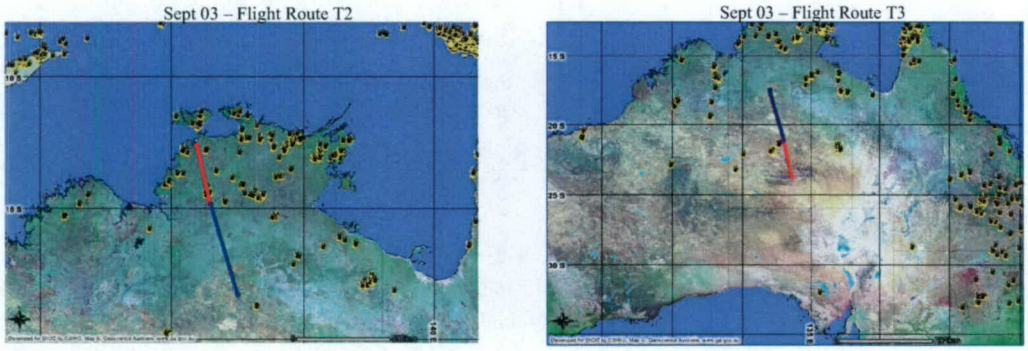

Figure 5: September 03 transit routes T2 and T3. The red line (shorter line segment) shows the location on the ground over which the 5 and 10 kft flight legs were flown. The blue line shows the same for the 15 and 20 kft legs. The black and yellow (white) markers indicate fire spots detected over the period 19-21 September 03.

Figures 4 and 5 show the flight paths for transit routes T2 and T3 and the number and location of fire spots detected for the period indicated. These images are taken from the Sentinel website [16] and permission has been given to reproduce them here. Sentinel Fire Mapping is a mapping tool designed to provide timely fire location data to emergency service managers across Australia. The mapping system allows users to identify fire locations that pose a potential risk to communities and property. Data is currently taken from the NASA Earth observation satellites TERRA and AQUA.

June 03 **Sept 03**

Figure 6: This figure highlights how much more burning is occurring across the continent as a whole in September during the period of the Adelaide to Darwin transit flights. The picture is essentially the same for the return legs from Darwin to Adelaide. The prevalence of bushfires along the East coast are likely to have an impact on the transcontinental measurements as the mean horizontal wind circulation at low altitudes (850 hPa) is strongly circulating across the continent (Figure 2).

Figure 6 is also taken from the Sentinel website and highlights how different the continental bushfire patterns are between the months of June and September 2003. Combined with the horizontal wind pattern maps (Figures 2 & 3) they provide a useful means for identifying how much smoke is likely to be dispersed across the continent.

3.1 Aerosol Loading

Data is available on the aerosol loading throughout the entire atmospheric column at different locations on the continent. The data is from ground based monitoring sites such as those operated by CSIRO with their network of ground based sun photometers that form part of the global AERONET aerosol network [17]. Satellites can also provide data on the aerosol loading globally and will give some indication of any major intercontinental aerosols that were present in the region during these transit flights. The satellite mounted Earth probe total ozone mapping spectrometer (TOMS) [18] is the first instrument that allows observations of aerosol across the land/sea boundary thus enabling us to observe desert dust storms, forest fires and biomass burning. The following information about TOMS is taken from the planet Earth science website [19]. Data from TOMS can be used to detect the presence of both ultraviolet (UV) absorbing aerosols and non-absorbing aerosols. The technique uses the ratio of the up welling radiance (or spectral contrast) between the 340 nanometer (nm) and 380 nm channels $\left[I_{340} / I_{380}\right]$. UV absorbing aerosols include smoke produced by biomass burning, black carbon from urban and industrial activities, agricultural dust, mineral dust coming from arid and semi-arid regions (desert dust) volcanic aerosols and ash. Carbonaceous aerosols generated by biomass combustion consist of a mixture of material with varying radiative properties; the absorbing fraction will contain elemental or graphitic carbon. Non-absorbing aerosols are primarily sulphate aerosols. UV spectral contrast is useful over both land and water because the UV reflectivity of these surfaces is low and nearly constant, unlike for the visible wavelengths. UV reflectivity of snow and ice, however, is high, therefore TOMS data is not as useful for detecting aerosols at high altitudes when snow and ice coverage produce high background surface reflection in the UV range.

The detection of aerosols from TOMS data involves a quantity called a residue. The N-value residue at 340 nm is defined as an aerosol index (AI) as follows:

$$AI = -100\left\{\log_{10}\left[I_{340} / I_{380}\right]_{meas} - \log_{10}\left[I_{340} / I_{380}\right]_{calc}\right\}$$

where I_{meas} = the backscattered radiance at that wavelength measured by the TOMS and I_{calc} = the model calculated radiance assuming an atmosphere of Rayleigh scatterers (pure molecular atmosphere) bounded by a Lambertian surface.

Non-UV absorbing aerosols result in a negative AI. For aerosol plumes at the most common height of 3 km, a TOMS aerosol index of less than 0.1 indicates a crystal clear sky with maximum visibility, whereas a value of 4 indicates the presence of aerosols so dense you would have difficulty seeing the mid-day sun.

The TOMS images shown in figures 7 and 8 are taken from the TOMS website and are reproduced here with the permission of the Laboratory for Atmospheres, Goddard Space Flight Center. For the periods of the transit flights during June 2003 there is no significant aerosol sources identified over the continent or being transported to the continent on global weather patterns. It appears that the concentration of aerosol produced through biomass burning in the Northern Territory is not significant enough to show up on the TOMS aerosol index. O'Brien and Mitchell [20] argue that this is not likely to be the case and that in fact to observe the prominent biomass burning smoke in the Northern Territory during the dry season simultaneous analysis of the TOMS AI data and AERONET sun photometer Aerosol Optical Depth (AOD) data from the Jabiru field station is required. Locally, fires were a major source of aerosol production as can be seen from the measured aerosol data and the fire maps.

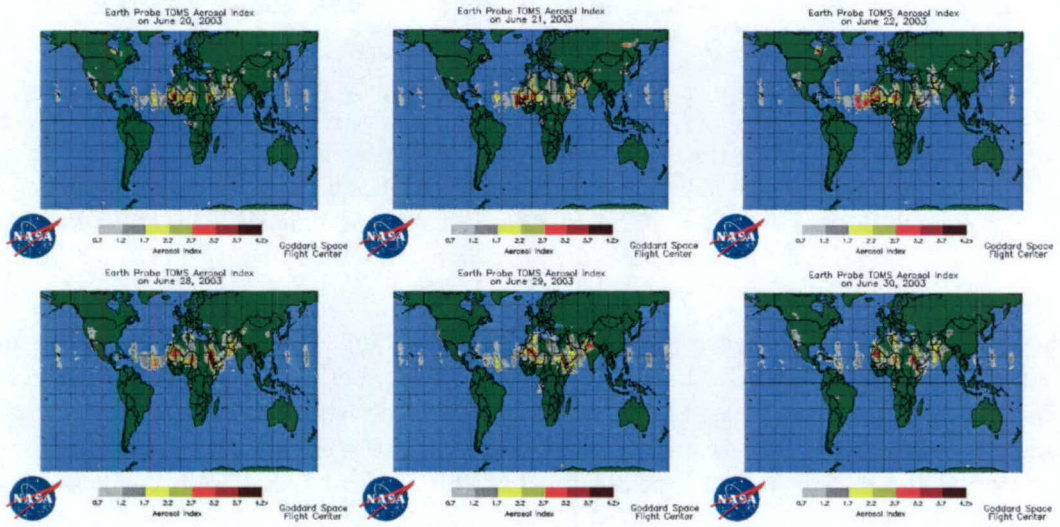

Figure 7: Images representing the UV absorbing aerosol loading over the globe as measured by the Earth probe TOMS during the time of the June transit flights. These images indicate that there were no major offshore (intercontinental) sources of aerosol being transported to Australia during this time.

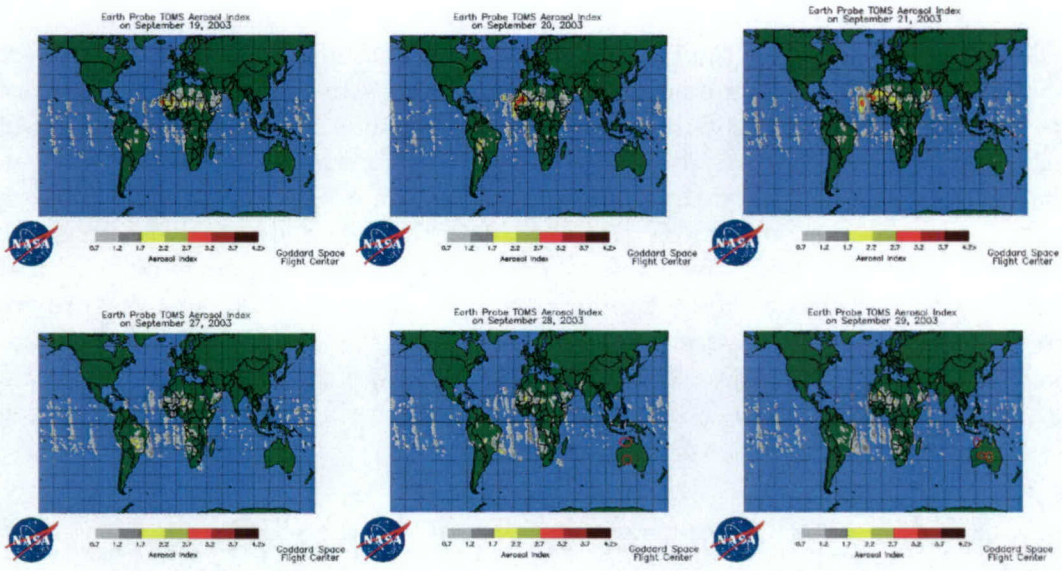

Figure 8: Images representing the UV absorbing aerosol loading over the globe as measured by the Earth probe TOMS during the time of the September transit flights. These images indicate that there was what appears to be most likely dust transported from Africa (and possibly biomass smoke from South America) over the Indian Ocean and relatively small amounts across Australia. (See in particular the last two images. The red circles highlight regions of aerosol.)

For September there is the clear presence of intercontinental aerosol over Australia (e.g. the red highlighted circles in the last two images for September). It is possible that some of the large concentration of coarse mode aerosol has come from these offshore sources during the return transit flight from Darwin to Adelaide in September. The peak in aerosol concentration as a function of altitude occurred at 5 kft (about 1.5 km), which is below the altitude where optimal TOMS aerosol detection occurs (3-5 km). The majority of this coarse mode aerosol is likely to be sea salt swept in land by low altitude winds coming from the South South West over South Australia and during the morning flight from Darwin to Alice Spring from the North North West over the Northern Territory.

We observed in ground-based measurements at Jabiru during this period that the coarse mode concentration peaks in the three hours or so to midnight and then falls off through the early hours of the morning as the winds start to move around from the East. During the morning flights there were still significant amounts of coarse mode aerosol in the atmosphere. Over the period of the four hours of the flights over Jabiru the ground-based coarse mode aerosol number concentration decreased. Significant amounts of sea salt were measured using a MOUDI sampler on the ground at Jabiru [1]. Each sample was over a 24 hour period. The one min samples collected with the GRIMM particle size counter at Jabiru [1] were used to determine the diurnal variation in fine and coarse mode aerosol number concentration during the campaigns.

The ground-based measurements at Jabiru during the period shortly after the T3 flight in September point to elevated levels of sea salt being present. The TOMS data indicates that

there was intercontinental aerosol over the continent during September and hence this is a possible source of aerosols

4. Aerosol Flight Chemistry

During the transit flights from Adelaide to Darwin only (transit routes T1 and T2) the filter unit was run. Particles were collected on filters to be weighed to gain an appreciation for how much aerosol mass density is likely to be present in the atmosphere. Two filters were used a lower and upper altitude filter. The upper altitude filter was used above 8000 feet. The filters collect aerosol via the isokinetic inlet and sampling line.

Table 2: Total measured mass density over the entire continent for low and high altitudes during June and September 2003

TRANSIT FLIGHT FILTER	TOTAL MEAS. MASS CONC. μG/M3 (± 3μG/M3)
June 2003 Adel-Darwin	
Low	9.5562
High	3.3225
September 2003 Adel-Darwin	
Low	11.3603
High	5.6158

Table 2 highlights just how little aerosol mass was in the atmosphere. These are exceedingly low levels particularly for the high filter. In fact for June high the error in the mass is as large as the measured mass. Determining individual inorganic ionic species from the samples enables us to assess what the basic chemical composition of the aerosol is. It is worth noting that in particular for the flights into Darwin a significant component of the mass will have come from organic carbon (OC) and black carbon (BC) (or soot) from the biomass smoke. A small amount of soil-based material may also contribute to the low altitude measured mass. The data we present below is only for the inorganic elements determined by ion chromatography. The fire maps (Figures 3 & 4) are obvious indicators for the location of fires and hence we are certain that smoke was a major constituent of the accumulation mode aerosol for transit routes T2 and T3.

The following charts show the elemental breakdown for the low and high filter samples for both June and September integrated over transit routes T1 and T2 (i.e. the entire continent). The upper atmospheric chemistry is most likely to be uniform over the continent so we do not lose much detail. The low altitude results will have included sea salt from Adelaide and Darwin but not much from Alice Springs. Integrating over the

entire continent removes information on the location of where major chemical species were collected but at least provides enough data to be able to identify major species.

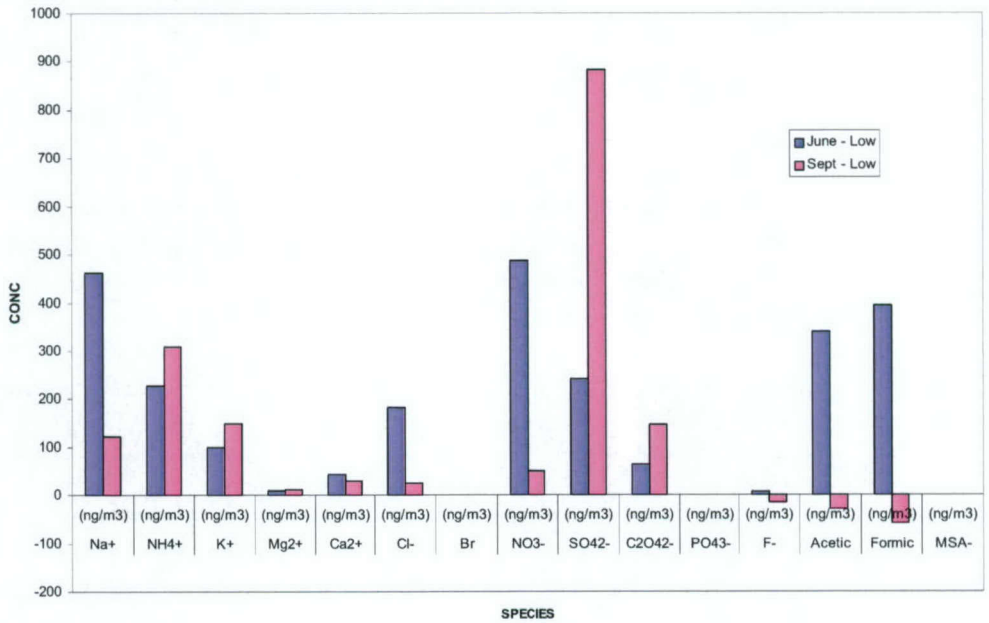

Figure 9: Individual ionic species mass concentration (ng/m3) for the transit flight from Adelaide to Darwin for the Low altitude filter (< 8 kft) for both June and September.

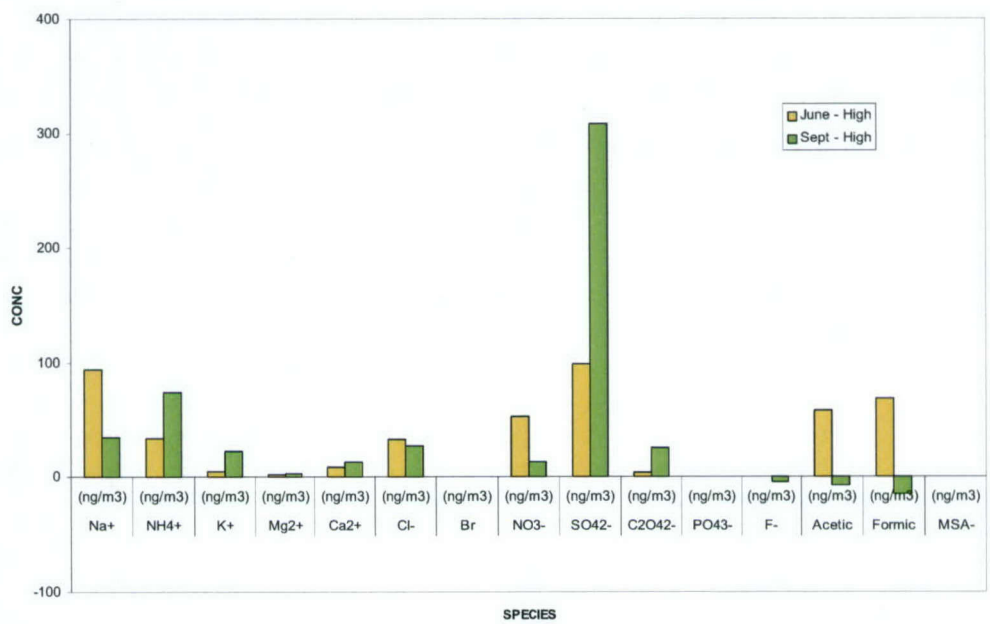

Figure 10: Individual ionic species mass concentration (ng/m3) for the transit flight from Adelaide to Darwin for the High altitude filter (> 8 kft) for both June and September.

The low altitude filter results reveal that sodium and chlorine are major contributors to the aerosol mass particularly for June. The larger amount of sodium and chlorine (sea salt) in June is not totally surprising, as it may have depended on the direction of the winds on leaving Adelaide and arriving in Darwin respectively. The coarse mode volume distribution in September for transit route T2 appears larger than for June (Figures 17 and 19). If we were to assume that most of the coarse mode aerosol was sea salt then the volume distribution and the chemistry data would appear to contradict each other. But we have to keep in mind that the chemistry data is integrated over the entire continent. Alternatively there may have been other major constituents that composed the coarse mode in September that were not present in as high concentration in June close to Darwin. We still observe high levels of sea salt in the high altitude filter as the artificial delineation of 8 kft between the low and high altitudes does necessarily correspond to the inversion height between the BL and the FT. If it did we would expect to see less coarse mode aerosol in the high filter. As it is the overall concentration of sea salt is lower in the high altitude filter.

Sulphate and ammonium compose much of the fine mode aerosol as secondary aerosol material formed by chemical reactions resulting in gas to particle conversion. Acetate and formate are tracers of biomass burning. The large values for June are most probably a result of flying directly through the cluster of fires just South of Darwin during the 5 kft flight leg. The result for September is unusual, as we would expect to see similar levels of acetate and formate as for June. These results may be due to experimental error due to working with such small amounts of mass. This is reflected in the negative values recorded for these species for September, which indicates that experimental error, and not the absence of these tracers is the cause of the unusual results. In any case we look to other information to confirm that biomass smoke was the dominant aerosol in September for transit routes T2 and T3 (Figure 5).

We have included some data on the chemical species mass distribution as measured on the ground at Jabiru. The aerosol was collected by a twelve stage Micro-Orifice Uniform Deposit Impactor (MOUDI) and subsequently inverted. Ion Chromatography was used to produce the mass distribution for each identified inorganic chemical species and BC levels were determined via a light absorption technique [1]. This highlights how different species are partitioned between the fine and coarse mode and we expect the same or similar for the transcontinental aerosol. These distributions correspond to the 24-hour period immediately after the transit flight between Adelaide and Darwin. This data should be a fairly accurate reflection of what the major aerosol was aloft during the transits into Darwin (transit route T2 for June and September). Certainly these results are typical and confirm what is already understood about aerosol formation and distribution in the atmosphere ([11], page 441).

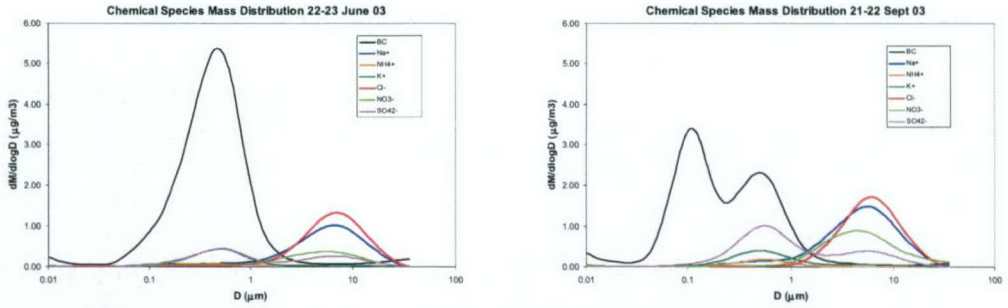

Figure 11: MOUDI mass distribution from Jabiru for a 24-hour period in June and September 03. The dominant BC peak for June is most likely due to a fire very close to Jabiru itself.

We can see from Figure 11 that BC dominates the fine mode (< 1 µm) whilst most of the sodium and chlorine are in the coarse mode. We find Nitrate and Sulphate in both modes as expected.

This data confirms at the very least that sea salt was present in the coarse mode fraction and that the significant amounts of sulphate and ammonium are consistent with the large accumulation peak observed particularly in September. We would expect the fine mode to be dominated by OC and BC coming from the biomass smoke.

5. Concentration and Size Distribution Data

Flight leg averages have been calculated for each transit route. These have been denoted T1, T2, T3 and T4. This improves the counting statistics and allows us to compare the magnitude and shape of the transit route distributions. Also displayed are the mean total number concentration and the count median diameter (CMD) for the SMPS and APS for each altitude. Throughout this chapter the error bars signify one standard deviation.

5.1 SMPS

5.1.1 June 2003

Total Concentration and CMD

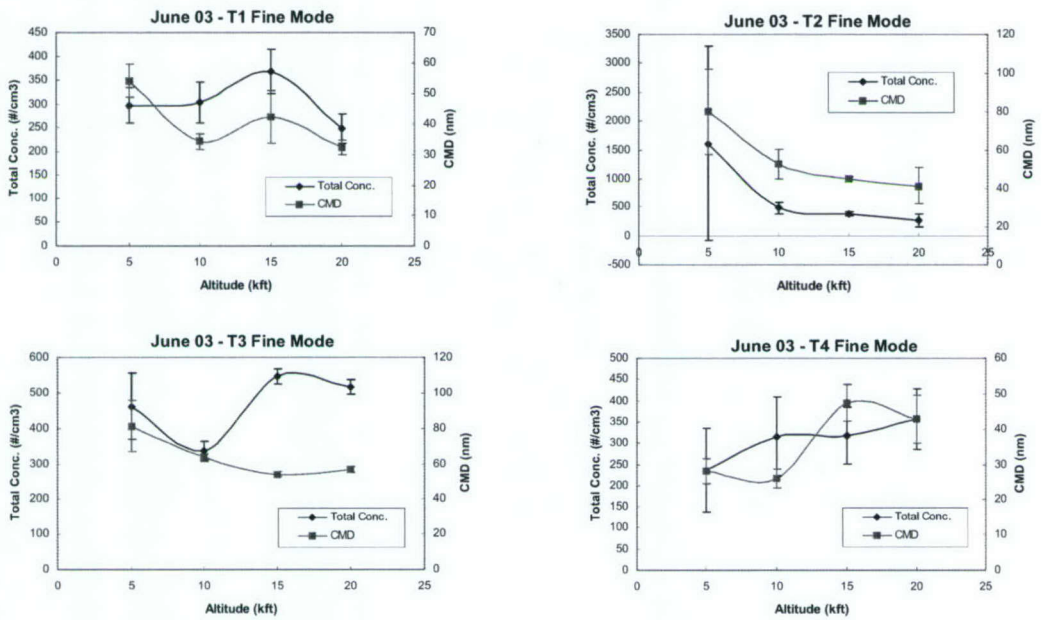

Figure 12: These plots highlight the variation in number concentration with altitude and also show the interrelationship between the concentration and the count median diameter (CMD). The error bars indicate ± 1 *std. dev. from the mean.*

The count median diameter (CMD) is the diameter (or bin), which has exactly half the concentration above and below it. If the total concentration increases and the CMD decreases this is an indication that there is a large number of smaller particles. Transit route T2 is dominated by the 5 kft leg due to the fires close to Darwin which extend to the 10 kft level (Figure 4). Comparing the T2 and T3 transit routes there is a larger total concentration at 15 and 20 kft for transit route T3 then for transit route T2. The CMD is also larger which indicates that much of this increase may be due to smoke from the fires identified in Figure 4. For transit route T3 the size distribution for these altitudes shows a dominate peak around 0.06 μm (Figure 13) which is the Aitken mode consisting of fresh young smoke from the fire to the West of the flight path (Figure 4). As the wind direction can change with altitude (Figure 2) it is possible during transit route T3 that the fires East of the flight path also impacted upon the 15 and 20 kft flight legs. Some of this smoke may have 'leaked' through to these higher altitudes. For transit route T2 there were a few fires to the West of the flight path for the 15 and 20 kft flight legs and the prevailing winds were

not blowing smoke towards the aircraft. Hence the overall concentration is down and so is the CMD indicating a prevalence of finer particles (more from the nucleation than the Aitken mode). This is consistent with the observed size distributions where there is an increase in concentration in the higher altitude flight leg distributions around 0.02 - 0.03 μm. This indicates that transit routes T2 and T3 appear to be primarily affected by biomass burning aerosol and any difference between the results for these transit routes can be explained by the location of fires with respect to the specific flight legs and the prevailing wind direction at that altitude.

The plots in Figure 12 allow a rapid assessment of flight legs. Flight legs with similar values for the total concentration and CMD indicate that the size distributions may be similar. If this is the case averaging can be performed over different flight legs to improve the counts without losing any detail on the vertical structure of the aerosol.

The most striking difference between transit routes T1 and T4 is the large CMD recorded for the 15kft flight leg for T4. Atmospheric conditions were extremely overcast on the approach to Adelaide (T4) with an extended cloud band between 5 and 10 kft. Analysis of the FSSP data reveals that there was cloud at the 5 and 10 kft altitudes but no evidence that the aircraft passed through cloud at 15kft (Figure 24). The larger CMD may have been a result of some scavenging above the cloud line resulting in broadening of the nucleation mode for transit route T4 compared with T3 (Figure 24) and resulting in an increase in the CMD. In other respects the results for T1 and T4 are similar indicating that the aerosol content of the air mass has not changed appreciably.

Size Distributions

The SMPS sampling time is 150 secs of which 120 secs are used to complete a full scan of the size range. There are insufficient samples per flight leg to provide a reliable conclusion about the statistical variation in counts for a given size bin across samples. The spatial distribution of aerosol in the atmosphere results in a Poisson distribution but additional sources of instrument noise can lead to a gaussian distribution. In this section we present the data for each flight leg. Averaging is performed over flight legs that are similar in both the magnitude and shape of the size distribution. This improves the count statistics without losing any information on the vertical profile of the aerosol.

Transit routes T1&T4

The average flight leg size distribution data for the Adelaide and Alice Spring transit routes (T1 and T4) show a large amount of variation in counts (concentration) between size bins. This is an indication that the data is noisy. The T1 5 kft size distribution for June 03 has a discernible bi-modal appearance. The two modes are a nucleation or Aitken mode and an accumulation mode. For the higher altitudes on this transit route there is only a single nucleation mode with the exception of the 10 kft leg which may have a small shoulder indicating an Aitkin mode as well. Certainly it is broader in appearance than the 15 or 20 kft flight leg distributions.

Transit route T4 is notable due to the absence of an accumulation mode for the lower altitudes. This is particularly the case for the 5 kft flight leg where the accumulation mode concentration is low compared with transit route T1. This probably reflects the fact that rain has washed out the majority of the accumulation mode at the lower altitudes below the cloud band. The 10 kft flight leg distribution is even broader with no discernible nucleation and Aitken modes. This may indicate that these two modes are overlapping producing what appears to be one large broad mode. In fact for this transit route we know that the aircraft passed through cloud for the 5 and 10kft flight legs. The fact that we are still able to discern a modal structure in the size distributions gives us confidence that the data is reflective of a true underlying size distribution.

The lower size bins exhibit the most variation between bins and for the same bin across samples. This most likely reflects the small concentration of this fine aerosol in the atmosphere for these transit routes and particularly for the higher altitudes we are testing the limits of performance of the SMPS.

We have chosen not to display the size distributions for these two transit routes, as the data is too noisy. For transit route T1 there was enough difference between the flight legs that we were unable to calculate vertical averages as was done for transit routes T2 and T3.

Transit route T2

Transit route T2 is dominated by the large accumulation mode peak in the 5 kft leg. This is due to the biomass smoke that the aircraft passed through during its approach to Darwin from the South. This smoke does not extend into the 10 kft leg. The inversion is lower in June (variable from day to day) and 10 kft was found to be in the FT. There are some differences between the size distributions at 10, 15 and 20 kft mainly in the total concentration with the shape remaining essentially the same (some slight broadening at lower altitudes). These differences may be the result of the fact that some smoke can 'bleed' through the inversion into the FT resulting in higher concentrations at the lower altitudes on approach to Darwin where fires started to become prevalent.

Figure 4 highlights this previous point with the 10 kft leg appearing to have been impacted on by the fires to the East. This results in a slightly broader distribution and higher concentration than for the 15 and 20 kft legs. The 10 and 15 kft distributions appear similar both in magnitude and shape. The 20 kft size distribution has a smaller peak and is less broad. For this reason we have decided to average over the 10 and 15 kft flight legs to improve the counts.

The distributions still show significant variation from size bin to size bin particularly at smaller diameters. However the basic shape of the size distributions is discernable indicating that the instrument has captured the essential underlying structure of the aerosol size distribution.

Transit route T3

Transit route T3 differs from T2 primarily due to the temporal shift of eight days and also because the higher altitude flight legs are flown closer to Darwin. In this case the 15 and 20 kft flight legs show a similar magnitude and shape with a large peak in the < 0.1 μm size range (Aitken mode or nucleation mode). The same applies to the 5 and 10 kft legs although the 5 kft leg shows a considerably broader accumulation mode probably due to the presence of higher smoke levels at lower altitudes emanating from one or more of the three fires in the vicinity of the flight legs (Figure 4). The winds being predominantly from the East and South East during June at these lower altitudes will have transported some of this smoke into the flight paths.

Another difference is that transit route T3 was flown during the morning when the atmosphere is more stable and the aerosol tends to stratify. Differences were observed between morning and afternoon flights during the measurements conducted over Jabiru. Examining the historical 9 am wind rose data for Australia [21] most weather stations report winds from the East to South East but interestingly enough essentially calm at Alice Springs. As these fires occur approximately at the same location (latitude) as the commencement of the 10 kft flight leg there would have to have been a Northerly component to the wind for smoke to have been observed at the 5 kft flight leg. Examining the 850 hPa (~ 5 kft) (Figure 2) wind circulation pattern for June we can see that the winds are blowing in a circular pattern over this region of the continent. This would have resulted in mixing of the smoke around this region at around 5 kft, which the SMPS was able to record at these lower altitudes.

The most striking observation is how much cleaner the air is in the South of the continent especially at the higher altitudes. This was most notable during the transit route T1 with very small concentrations of coarse mode aerosol measured (Figure 16).

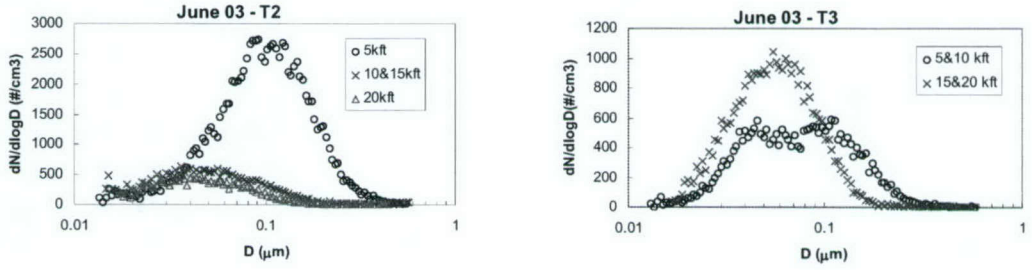

Figure 13: Flight leg size distributions for the two transit routes flown in the North of the continent. 5 & 10 kft, 10 & 15 kft and 15 & 20 kft signify an average has been taken over those two flight legs. The data shows variation bin to bin but the basic shape of the distributions is clearly discernable. In particular the clear bi-modal shape of the 5& 10 kft distribution for transit route T3.

5.1.2 September 2003

The SMPS experienced an operational problem on the return transit from Darwin to Adelaide in September. This problem was unable to be diagnosed and fixed within the limited time and space constraints associated with operating on an aircraft. As a result there is no data on the fine mode aerosol except what is available from the ASASP and the tail of the APS data. We also have data on the fine mode that comes indirectly from the nephelometer (Chapter 6).

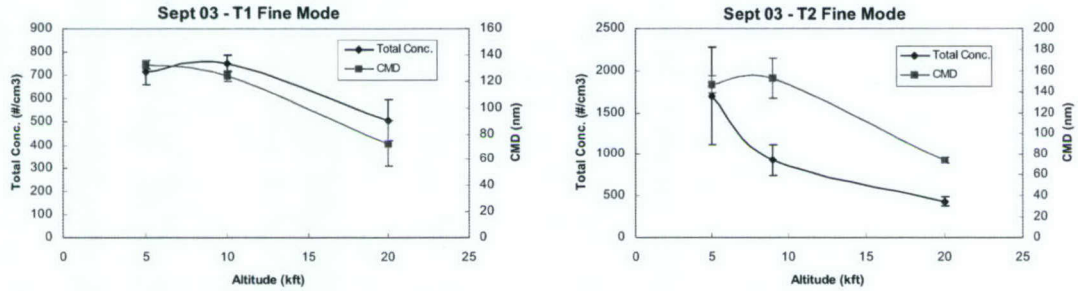

Figure 14: These plots highlight the variation in number concentration with altitude and also show the interrelationship between the concentration and the count median diameter (CMD). The error bars indicate ± 1 *std. dev. from the mean.*

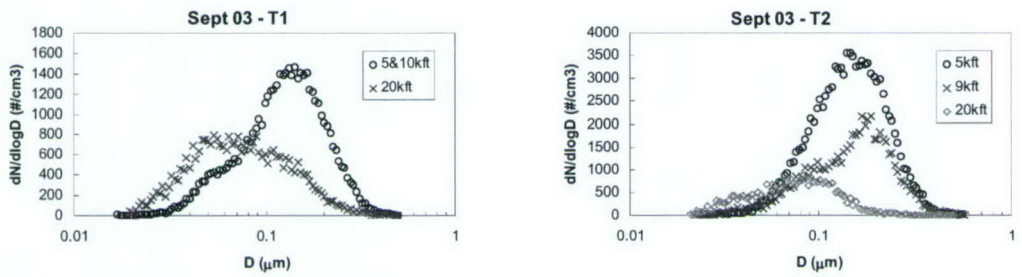

Figure 15: Flight leg size distributions for the two transit routes shown. 5 & 10 kft signifies an average has been taken over those two flight legs. The data shows variation bin to bin but the basic shape of the distributions is clearly discernable.

The total fine mode concentration is higher in September then it is for June but the most striking difference is the much larger CMD. This reflects most notably the large accumulation mode peak at 5 kft for transit route T1. The CMD remains large for the 10 and 9 kft flight legs (Figure 14). We observe that the total concentration at 10 kft is much smaller in June then it is for September. This indicates that the atmospheric conditions are considerably different between the two months. The inversion height occurs at a higher altitude in September. This is consistent with the results observed during the measurements conducted over Jabiru for the week in September. The 10 kft flight leg in all

cases was in the BL [1]. The CMD drops noticeably in the FT (20 kft) as expected since the accumulation mode is much smaller and the Aitken or nucleation mode becomes more significant.

For transit route T2 the shoulders in the size distributions around 0.07 μm at 5 and 9kft are most probably a result of a secondary peak due to a real Aitken mode. This is likely to be fresh young smoke. The large accumulation peak will be from more aged smoke and is clearly dominating in all cases. For transit route T1 the shoulder occurs at a smaller diameter around 0.05 μm and is probably more of a nucleation mode. It is possible that this is smoke from bushfires on the East coast of Australia. In fact it is similar to the 9 kft size distribution for transit route T2. There was considerably more burning occurring around the continent in September compared with June (Figure 6). Most of this is concentrated along the East coast and the top end of the Northern Territory with a few sparse fires to the West. The strongly circulating horizontal winds at 850 hPa (Figure 2) during September will have transported this smoke from the East coast out over the pacific and then back across the North of the continent circulating down through the centre. This would have resulted in smoke being present on the low altitude flight legs approaching Alice Springs and would explain the relatively large accumulation mode peak for this transit route.

Unfortunately we do not have SMPS data for the return transit routes T3 and T4. It is likely, based on the fire maps, that high levels of smoke would also have been recorded. The main difference between the return transits is that the low altitude flight legs are flown into Alice Springs and Adelaide. In the absence of any SMPS data we can look to the APS data for sizes less than 1 μm (Figure 19), which will tell us something about the tail of the accumulation mode and hence something about the nature of the aerosol. We also have the ASASP data, which extends further up the peak of the accumulation mode down to around 0.3 μm (Figure 25).

5.2 APS

5.2.1 June 2003

Total Concentration and CMD

The large number of submicron-sized particles dominates the total concentration from the APS. The noise due to the uncertainty in the large size particle bins becomes less of an issue for the total concentration. For this reason whilst the size distribution can itself become too noisy to display the total concentration does provide some information on the variation of the coarse mode over different altitudes and transit routes. The major difference between the SMPS and APS results is that the SMPS shows variation between two and potentially three separate modes (nucleation, Aitken and accumulation) whilst the APS only accounts for one complete mode (the coarse mode) and the tail end of the accumulation mode at lower altitudes. This is reflected in the CMD, which tends to remain

constant for the APS flight legs over the different transit routes but changes noticeably with the SMPS flight legs as the accumulation mode or the Aitken (nucleation mode) dominates the distribution. For the higher altitudes we do not measure a clearly defined coarse peak in the volume distribution.

The values for the coarse mode concentration are conservative as the APS counting efficiency below about 0.7 μm is less than one and so the concentration of these size particles is underestimated. Also larger size particle losses through the isokinetic inlet will affect the results. The total concentration and CMD are a useful way to summarise changes in the gross properties of the coarse mode aerosol across the continent.

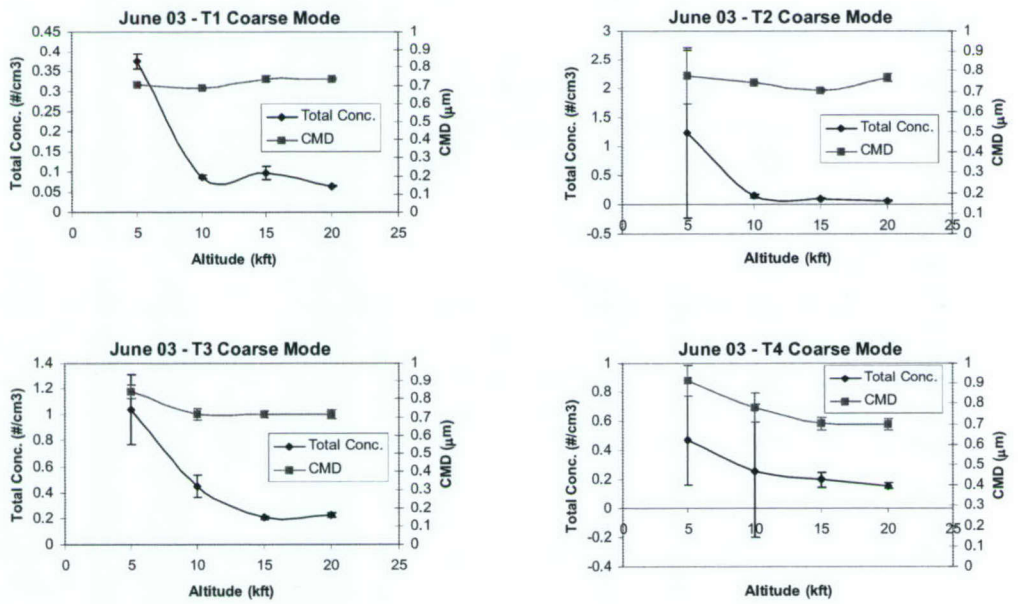

Figure 16: Total Concentration and CMD for the coarse mode aerosol (& tail of the accumulation mode) for June 03

The results for June 03 transit route T4 is impacted upon by the presence of cloud samples in the flight leg averages. This is particularly noticeable for the 10 kft flight leg with the large variation in the number concentration, which reflects the fact that only some samples were through cloud. Hence the T4 lower altitude results should not be taken as indicative of the actual aerosol loading in a clear atmosphere on approach to Adelaide.

Volume Distributions

For June we observe differences between the Adelaide to Alice Springs and Alice Springs to Darwin transit routes most notably at lower altitudes. This is presumably due to the tail of the accumulation mode resulting from biomass smoke. There are also differences between the transit routes (T1 and T4) and (T2 and T3) presumably because of the

different air masses being sampled eight days apart and due to the fact that the flight legs are over different terrain for the outgoing and incoming transits eg the T1 5 kft leg is approaching Alice Springs whereas the T4 5 kft leg is approaching Adelaide.

The coarse mode volume distributions show a large amount of variation bin to bin for large diameters due to the lack of coarse mode aerosol aloft and the sampling losses through the isokinetic inlet. Ideally we would like to be able to average over T1 and T4 and T2 and T3 flight legs to gain an additional improvement in the large particle counts. However the differences observed between the outgoing and incoming transits means we should proceed with caution so that we do not lose information we have on the aerosol size distribution variations both in space and time.

Transit routes T1 & T4

The coarse mode concentration is exceedingly low with average concentrations being around 0.08 (cm^{-3}) in the FT and as high as 0.376 (cm^{-3}) for the 5 kft leg in the BL. The concentration includes particle sizes down to 0.487 μm. After averaging over the 10, 15 and 20 kft there is no discernable coarse mode peak in the volume distribution. The 5 kft leg distribution has a more definite shape but it appears random above 3 μ m.

There is an insufficient number of counts in the large particle size bins to allow a useful volume distribution to be observed. This is an indication that the air is very clean and the instruments were not sampling very high levels of coarse mode aerosol on the approach to Alice Springs. Of all the transit routes flown in June and September this one showed the lowest levels of total number concentration across the entire size range observable with the instrumentation.

The coarse model concentration is larger for the T4 transit route then was the case for transit route T1. The 5 and 10 k ft flight legs show a large coarse mode as a result of the presence of cloud. This is reflected in the total concentration which shows large variation between samples, some including cloud others not. For these reasons we have chosen not to show volume distributions for transit routes T1 and T4.

Transit route T2

There is a large drop in concentration between the 5 kft and the 10 kft flight legs. The concentration at 10 kft for this transit route is the same magnitude as at 5 kft for transit route T1 indicating the air is more loaded with aerosol. The concentration drops to 0.102 (cm^{-3}) at 15 kft and to 0.061 (cm^{-3}) at 20 kft. There is more of a shape to the coarse mode size distribution for this transit route but for higher altitudes there is an insufficient number of counts in the larger size bins to conclude that there is a definite coarse mode with a peak and an associated spread. The 5 kft leg has a larger concentration around 1.23 (cm^{-3}). The volume distribution for this flight leg is shown in Figure 17.

Transit route T3

The volume distribution for the 5 kft flight leg shows a clear coarse mode peak centred at 3 μm (Figure 17). It is similar to that measured during transit route T2 at the same altitude. However it is much larger than the coarse mode that was measured eight days early on transit route T1 when approaching Alice Springs. This indicates how much the aerosol can vary over time and in particular for the lower altitudes how dependent the coarse mode concentration is on local events. Both the 5 kft flight legs for transit routes T1 and T3 are over similar regions of the continent albeit South and North respectively of Alice Springs.

The ground level winds are more from the North in South Australia becoming calm at Alice Springs (9am wind rose data, [21]). In generating coarse mode aerosol via abrasive contact with the surface the direction of the wind should not matter. From the wind rose data the Southerlies North of Alice springs are stronger than the Northerlies South of Alice Springs and hence this may have resulted in different coarse mode aerosol levels. It's difficult to make any more comments without additional information.

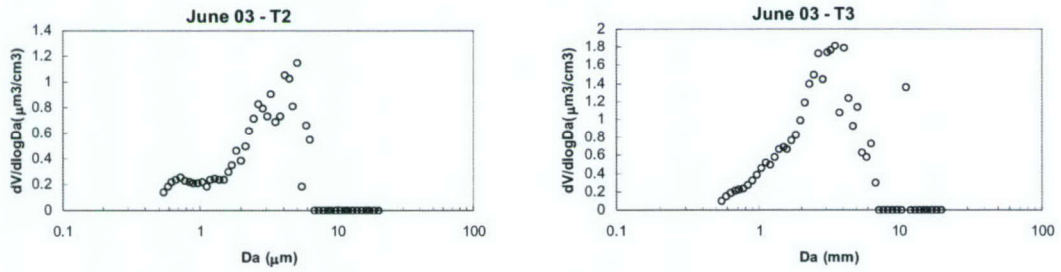

Figure 17: Volume distributions for the 5kft flight leg. Count variation bin to bin is indicative of noise and reflects insufficient samples (counts vary within a bin from sample to sample a lot) for accurate statistical representation of the distribution. However it is quite clear that there is a peak around 3 μm associated with the course mode.

5.2.2 September 2003

Total Concentration and CMD

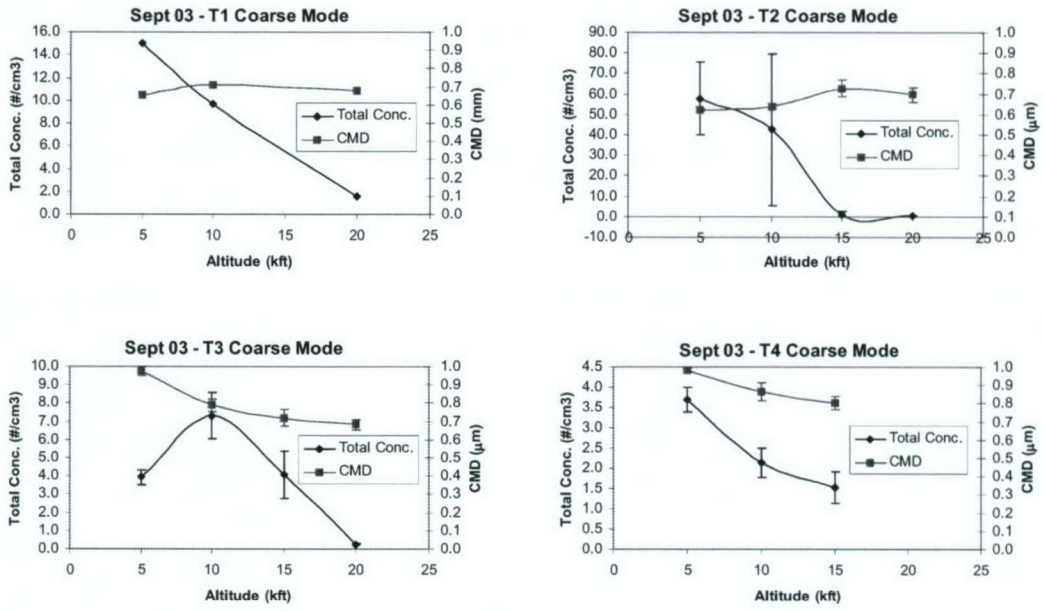

Figure 18: Total Concentration and CMD for the coarse mode aerosol (& tail of the accumulation mode) for Sept 03.

Problems with the data logging laptop during transit route T4 meant that data was not being logged for the 20 kft flight leg.

There is a notable difference between the information conveyed in Figure 18 and that of the volume distributions (Figure 19) particularly between the 5 and 10 kft flight legs for transit route T3. This is mainly due to the fact that the total concentration includes the size bin < 0.523 µm that in the case of the 10 kft flight leg dominates the number concentration. The APS logs particle size < 0.523 µm for total concentration but not for the size distribution. The volume distribution weights in favour of large sizes due to the D^3 dependence and hence we see that the distribution of large size particles dominates in the 5 kft leg over the 10 kft. This is despite the fact that if we include the <0.523 µm bin the total number concentration is double for the 10 kft leg. It is likely that the increase in concentration for this bin at 10 kft is due to the fires that are in the vicinity of the lower altitude flight legs on the T3 transit route, contributing to a large accumulation mode tail. We have chosen to include this bin in the total concentration plots, as it is still valid for a relative comparison of different flight legs for a given transit route and between transit routes. However when looking at the total magnitude of particles in the coarse mode it

may be better to remove this bin from consideration. We have left out this bin in the display of the volume distribution, which will not affect the results greatly due to the D^3 dependence.

The large variation for the 9kft flight leg of transit route T2 is due to one extremely large sample, which is most likely a result of the aircraft flying directly through a smoke plume. For this one sample the total concentration was 237 (cm^{-3}). This also increases the average concentration for the 9kft flight leg. This is the most likely explanation as to why the 9 kft flight leg coarse mode size distribution is larger in magnitude than the 10 kft flight legs. The altitude difference of 1000 ft may have some impact on this but should not be significant.

Volume Distributions

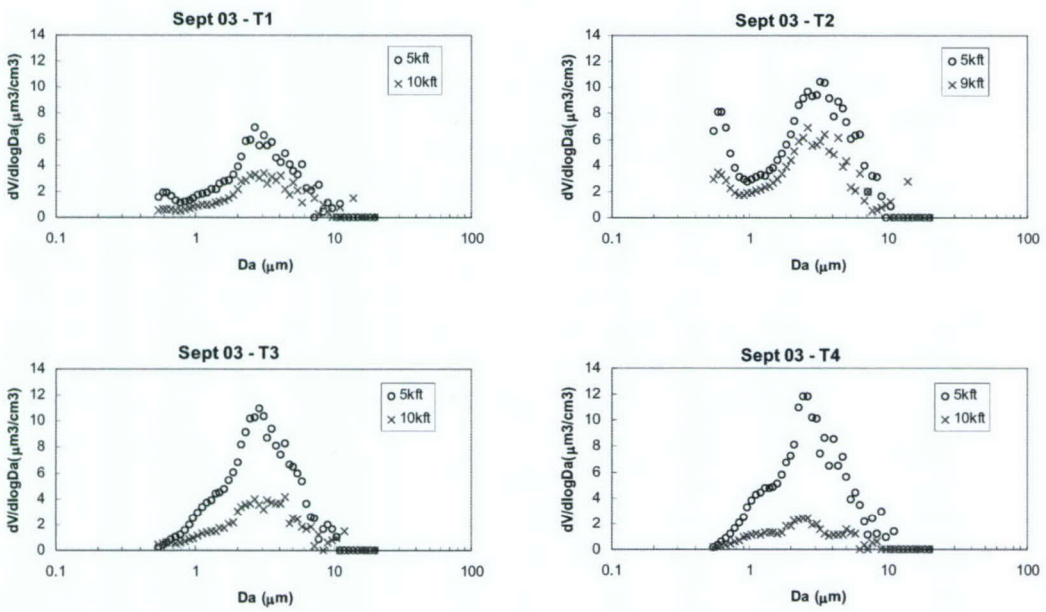

Figure 19: Coarse mode volume distributions for the BL flight legs for all four transit routes

The coarse mode peak is much more pronounced and persists at higher altitudes for the September transit flights than was the case for June. The smaller size bins for the outgoing transit routes T1 and T2 show an increase below 1 μm. The decrease in concentration that occurs for the last few bins is due to counting inefficiencies with the APS and is not due to the underlying shape of the size distribution. This contrasts with the incoming transits T3 and T4, which show a sharp drop off below 1 μm with no increase at all even in the last few size bins. The larger increase in the size distribution below 1 μm for transit route T2 is due to biomass smoke and the APS is responding to the tail end of the accumulation mode

associated with this biomass burning smoke. The shoulders in the T3 and T4 5 kft size distributions tend to indicate the presence of two coarse modes one peaking at 1-1.5 μm and the other at about 3 μm. The presence of a shoulder is quite common in the SMPS distributions (Figure 15) and indicates the presence of two modes, the Aitken mode due to young fresh smoke and the accumulation mode associated with aged smoke (more than a few hours).

If we examine the historical 3 pm wind rose data for the continent [21] and compare June with September (winter compared with spring) we observe that winds are more frequently from the South South West and stronger in magnitude in September. This is likely to have generated and transported a large amount of sea salt inland and hence is the most likely explanation for the larger coarse mode in September compared with June for the T4 transit route. Without more localised data on the chemical composition of the aerosol it is difficult to conclude what the two coarse modes are likely to be. We know sea salt will be present and the rest may well be wind-blown dust that could potentially have been transported on intercontinental weather patterns (Figure 8).

It is interesting to examine further the difference in the volume distributions for sizes less than 1 μm. The smaller increase in the distribution less than 1 μm for transit route T1 is expected as the amount of smoke in the atmosphere due to the East coast fires will have been much less than that due to the top end fires, which impacted on transit route T2. The curious result is that the APS is picking up little if any smoke for transit routes T3 and T4 despite the continued prevalence of bushfires on the East Coast. The volume distributions show a drop off to effectively zero indicating no accumulation mode at all. Unfortunately we do not have any SMPS data to confirm this result. We can look to the ASASP however (Figure 25). On comparing transit routes (T1 & T2) with (T3 & T4) we observe that the lowest ASASP size bin concentration is much lower for transit routes T3 & T4. In fact the concentration is well over 1000 (cm^{-3}) for T1 and T2 and drops down to under 500 (cm^{-3}) for T3 and T4 and closer to 100 (cm^{-3}) for T4. The level for T2 reflects a large amount of smoke around Darwin as expected. This is consistent with the APS volume distributions and indicates that the accumulation mode for transit routes T3 and T4 was much lower. The nephelometer Green scattering coefficients confirm these low level of aerosols. For these transit routes the scattering coefficient is in the 10^{-6} s compared with 10^{-5} s for transit routes T1 and T2 at the lower altitudes where we would expect to see accumulation mode aerosols from smoke dominating. All instruments are pointing towards one fact, for these flights the accumulation aerosol was not as significant as expected.

The reasons for this are not completely understood. It is unlikely that if the winds were transporting smoke as for transit routes T1 and T2 that we would have obtained such a low level for the accumulation mode. It may be that anomalous wind patterns were the norm for that day (Figure 3) which would explain why smokes levels around Alice Springs were not as high for T3 compared with T1. Separate wind direction data from the aircraft if available would confirm if these anomalous wind patterns were occurring during these transit flights. This would have resulted in smoke from the East coast fires

being blown out over the Pacific with no circulation back over the continent and hence no recording of smoke during the T3 and T4 transits, as we observed.

These results just highlight how aerosol levels in the atmosphere can vary on any given day depending on weather patterns and the prevalence of aerosol sources.

5.3 ASASP and FSSP

The FSSP recorded low levels of coarse mode aerosol aloft particularly for June. This was observed in the APS data with no counts recorded for sizes larger than 7 μm. The FSSP is sensitive to particles in the size range 4 - 50 μm with the lowest geometric mean diameter (GMD) at 5.4 μm and the next at 8.4 μm. The concentration measured is consistent with APS levels. In this case there is no value in using the sparse FSSP data unless we want to fit volume distribution functions to the coarse mode size distribution data. Then the FSSP should be used to improve the tail of the size distribution. For completeness we present the FSSP data for June.

The ASASP spans both the accumulation and coarse modes but neither completely. It is an additional source of data that can be used to assess the validity of the SMPS and APS data. If we find good agreement between these three sizing instruments all employing different measurement techniques, then that will improve our confidence in the results. Again, in particular for June, the counts are very low for the coarse mode.

Refer to the next section and Appendix A for more detail about the ASASP and FSSP size distributions.

5.4 Combined Differential Size Distribution

Parts of this data should be viewed with care as it is obtained at the very limit of instrument sensitivity and the levels in particular of coarse mode aerosol aloft in the atmosphere especially in clear continental air mass is exceedingly low above a few thousand feet.

All of the size distribution data has been converted from raw counts to number concentration per bin divided by the logarithm of the ratio of the bin upper and lower limits. Because the instruments provide sizing through different means (section 2.1) we have used a generic D to symbolize diameter, which for the SMPS is the particles physical diameter, the aerodynamic diameter for the APS and the particle equivalent optical diameter for the ASASP and FSSP. The APS has not been converted to physical diameter, as we do not have sufficient data to determine the aerosol density for the coarse mode. At higher altitudes the APS also suffers from a pressure induced size shift [1], which results in particles being sized smaller than they actually are (in aerodynamic terms). The result of applying the density correction and pressure induced size shift correction for the higher altitudes are likely to offset each other and hence what we have displayed may be close to reality. For the lower altitudes the pressure induced size shift is less important and the

density correction would result in the distribution shifting to the left, the amount depending on the magnitude of the unknown aerosol density.

The diameter of the particles as measured by the probes (optical equivalent diameter) is a function of particle refractive index and particle shape. Characterisation of particle shape is very tedious requiring electron micrograph analysis of particle samples collected on board the aircraft. Refractive index calculation is less tedious but requires sufficient chemistry data to be collected to allow for input into a thermodynamic aerosol model such as Scape II [22] to determine an average aerosol refractive index and density. The mass of material collected on the transit flights is too small in magnitude to allow for this analysis and hence we are unable to determine what the aerosol refractive index was as a function of ambient conditions. The calibration of the probes using spheres (glass beads) of a known refractive index is likely to introduce some error into their sizing accuracy. Shape effects will also have an effect on both the APS and the probes to varying degrees but without knowledge on the shape factor are hard to quantify especially in the case of the probes, which make the assumption of scattering from spheres.

Presented in this form the data displayed in Appendix A shows clear mismatches that can be explained as a result of the above effects. The data however shows reasonable agreement in many cases given that no corrections have been applied. Each instrument has captured the essential nature of the underlying size distribution. If this data were to be used to fit theoretical size distribution functions then these corrections would need to be applied to allow for a smooth fit. In this case the preferred method is to fit each mode separately [1]. Some of the more interesting features of these size distributions will be discussed in Appendix A.

The ASASP encountered some operational problems during the June campaign. As a result it was not performing optimally. The data therefore is not to be considered as reliable as for September. The other instruments were performing correctly and hence the SMPS and the APS data should be considered the most accurate for June. The FSSP data in June is generally zero or exceedingly low and hence is of limited value.

As a comment and to some degree a justification for presenting this uncorrected size distribution data for all available flight legs for all transit routes we mention the following points.

1. We thought it was better to display all size distribution data than to highlight only the 'best' data.
2. The effort involved to correct the data for each flight leg is significant for what are essentially data of opportunity.
3. Practically to achieve point 2 would be difficult if not impossible due to the extremely small mass of material in the atmosphere over the continent and hence collected on filters for chemical analysis.

4. It allows us to illustrate that in a majority of cases the instruments have captured the shape and to a lesser degree the magnitude of the underlying particle size distribution.
5. In general the agreement at the uncorrected level is good, which indicates that the data is real, and not an anomaly of one particular sizing instrument.

We accept that this may leave us open to criticism. Ideally in the future if knowledge about the aerosol density and refractive index becomes available then corrections to the APS and probe size distributions should be applied. This is particularly true if this data is to be compared with future measurements and or if it is to be used to fit theoretical size distribution functions. In this case careful attention should be paid to weighting the data in terms of quality. This is particularly true for the coarse mode tail where particle concentration is being measured at the absolute limit of the APS and to the limit of the FSSP. To some degree if agreement between the SMPS and APS at their respective tails can be achieved then once size distribution functions are fitted to the separate modes these theoretically fitted functions can provide the tail of the accumulation and coarse modes. All these individual flight leg combined size distributions are presented in Appendix A along with discussion.

6. Scattering Coefficient Data

The results for June and for September are shown as modified box plots for each transit route. Caution is required when viewing these box plots for each transit route since the number of samples for the flight legs is not necessarily constant. For the higher altitudes where the averaging time was generally 4 or 5 times longer than at the lower levels there are fewer samples. This only becomes an issue for September.

Angstrom Exponents (450/700 and 450/500)

The nephelometer photomultiplier tube (PMT) sensitivity for blue and red wavelengths is not as good as it is for green. In particular the red PMT gain is set high at the expense of increasing the dark signal. The trade off between increasing the gain to improve sensitivity whilst limiting the noise means there is a finite limit as to how much the sensitivity can be increased. The result is that the green PMT is the most sensitive under normal operating conditions [1]. However the blue and red signals do provide some information on the wavelength dependence of the scattering of visible light by aerosols. This information can be used to gain an appreciation for the nature of the underlying size distribution independently of the separate measurements coming from the sizing instruments. Rather than overload the reader with temporal traces of the scattering coefficient for all three wavelengths at each altitude for each transit route we have chosen instead to present modified box plots of the green scattering coefficient and the angstrom exponent for each flight leg.

The angstrom exponent is defined as

$$\overset{o}{A}(\lambda_B / \lambda_R) = \log(\beta(\lambda_B)/\beta(\lambda_R))/\log(\lambda_R / \lambda_B)$$

where $\beta(\lambda_i)$ is the scattering coefficient at the wavelength λ_i, $i = B, R$ and with λ_i given in nm.

The green scattering coefficient in some sense represents the mean amount of scattering that takes place in the visible spectrum and the angstrom exponent summarizes the variation about that mean wavelength. For example we expect large particles to scatter a greater amount of red light and in particular fine smoke particles will scatter predominantly at a blue wavelength. The angstrom exponent being a difference in the scattering coefficient on a log scale normalised to the log of the wavelength interval reflects an increase or decrease in either fine or coarse mode particles between measurements. This would be harder to ascertain by simply viewing the temporal traces of the scattering coefficients for the individual wavelengths.

The angstrom exponent can be used to determine what the power exponent is for the underlying size distribution if one assumes that a power law (Junge) form of the size distribution is adequate to characterize the aerosol. Rather than investigate this line of reasoning further we have chosen to simply use the angstrom exponent as a means of estimating the relative contribution of fine and coarse mode aerosol to the scattering. We prefer to use the data measured directly from the sizing instruments to determine the nature of the size distribution. The comparison of the total concentration of the fine and coarse modes from the sizing instruments with the nephelometer data is a way to determine the consistency of the results coming from the two types of instruments. We cannot determine exact amounts of fine and coarse mode aerosol from the nephelometer data however we can use the angstrom exponent to get an indication as to the relative abundance of the two modes and more particularly, as mentioned, to identify changes between flight legs and or transit routes.

The 16 and 20 kft flight legs for transit route T2 in September should be viewed with care. The red scattering coefficient for the 16 kft leg is on the detection limit of the instrument. It is just below the detection limit for the 20 kft leg. The blue scattering coefficient is above the detection limit for both flight legs so we have chosen to calculate angstrom exponents for these two flight legs.

6.1.1 June 2003

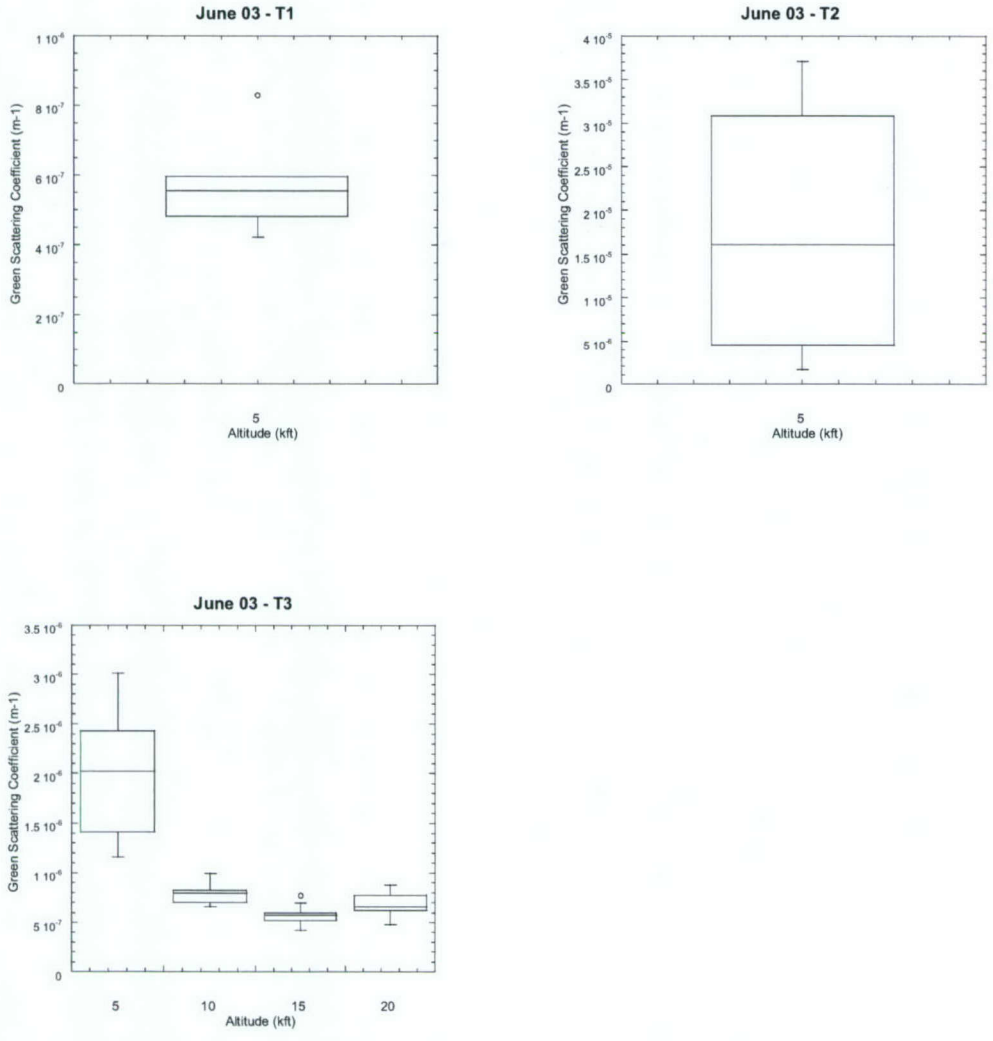

Figure 20: Modified Box plots of the green scattering coefficient for each transit route of the transit flights between Adelaide and Darwin for June 2003. The scattering coefficients are given in units of m^{-1}. The solid line is the median value and the lower and upper line of the box represent the 25th (lower quartile (LQ)) and 75th percentile (upper quartile (UQ)) respectively. The whiskers are points within which data is statistically significant i.e. the upper whisker = UQ + 1.5 IQD and the lower whisker = LQ - 1.5 IQD, where IQD= UQ - LQ is the inter quartile distance. The open circles represent outliers. Flight leg/ Transit routes without any data are below the green wavelength detection limit.

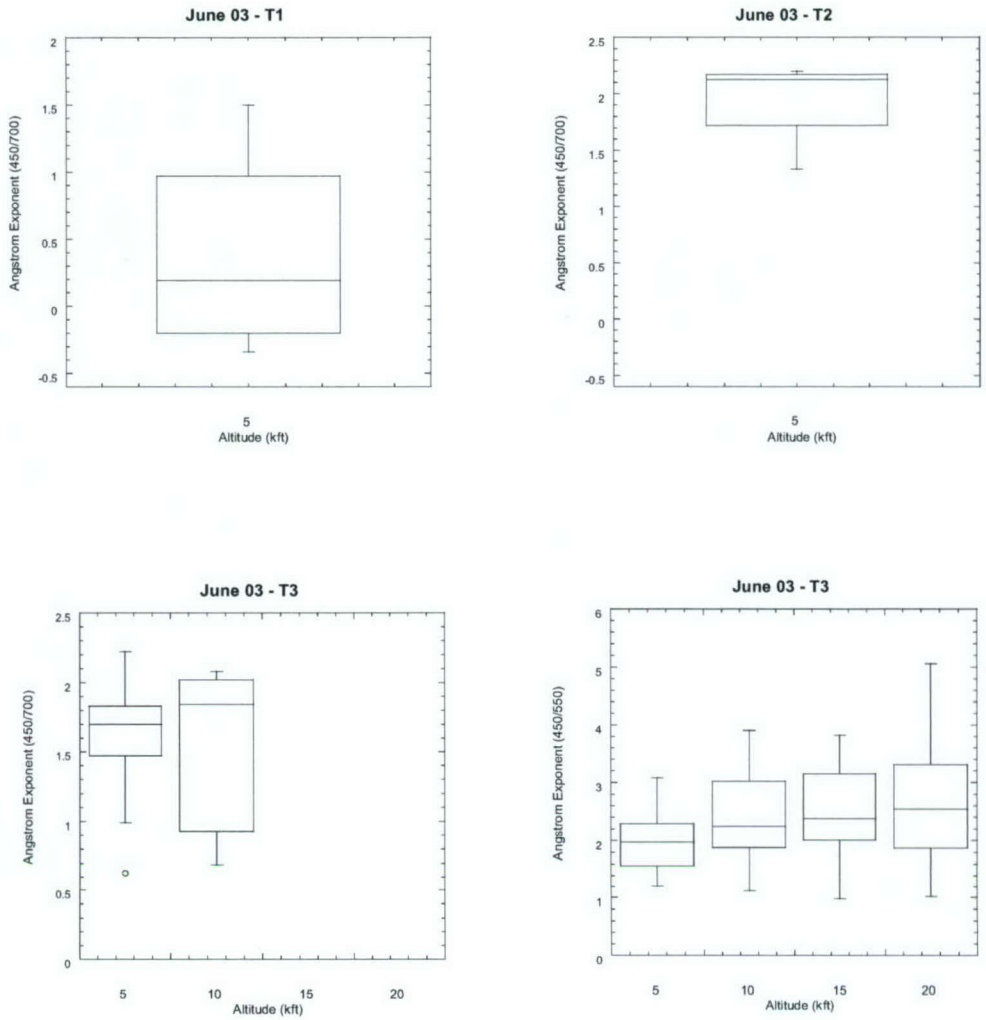

Figure 21: Modified Box plots of the Angstrom Exponent $\overset{o}{A}$ (450/700) for each transit route of the transit flights between Adelaide and Darwin for June 2003. Flight legs without any data are below the red wavelength detection limit. The final plot is of the Angstrom Exponent $\overset{o}{A}$ (450/550) calculated using Blue and Green scattering coefficients for transit route T3.

The most notable feature of the June transit flight between Adelaide and Darwin was just how clean the air was. Even the flight between Alice Springs and Darwin showed a noticeable scattering signal only at 5 kft. The return transit between Darwin and Alice Springs showed the largest scattering coefficients in all three wavelengths. For this transit route (T3) at all altitudes the blue and green scattering coefficients are above the detection threshold but still exhibit variations between samples. The red signal is most likely just

below the detection threshold and therefore cannot be considered reliable. For the other three transit routes only the 5 kft flight leg for transit routes T1 and T2 shows a real scattering signal while the signal at the other altitudes lies below the detection threshold of the instrument [1].

The interesting difference between transit routes T2 and T3 is that there is very little scattering taking place above 5 kft for transit route T2. The 5 kft median green scattering coefficient is larger for transit route T2 since this leg was flown through the cluster of fires just South of Darwin. This was also reflected in the extremely large accumulation mode peak in the SMPS size distribution. The peak is smaller for transit route T3 as discussed earlier since the number of fires having an impact on the instruments on the approach to Alice Springs was far less. Conversely the higher altitude (15 and 20 kft) size distributions for transit route T3 are much larger then for transit route T2. This indicates a greater prevalence of fine mode particles less than 0.1 μm, which shows up as a large peak in the SMPS size distribution for transit route T3 15&20 kft flight leg average. It is reflected in the scattering coefficients by a larger blue scattering signal than green and the red scattering signal is below the detection threshold of the nephelometer. This just confirms that the fine particles do not scatter the longer wavelength radiation as much. In the case of transit route T3 we can calculate the angstrom exponent at $\overset{o}{A}(450/550)$, using the blue and green scattering coefficients, for these higher altitudes.

The 10 kft flight leg for transit route T2 has a peak in the SMPS distribution that is similar in magnitude to the 10 kft flight leg for transit route T3 but is possibly centred at a slightly smaller diameter. This is the most likely explanation as to why for transit route T3 the red scattering signal is just on or above the detection limit of the instrument but falls below it for transit route T2. Again this means we can calculate the angstrom exponent at $\overset{o}{A}(450/700)$ for the 10 kft flight leg for transit route T3 but not for transit route T2. Also the smaller peak in the SMPS size distribution for higher altitudes (above 10kft) produces less scattering at blue and green wavelengths and these signals remain below the detection limit of the nephelometer. This remains the case for all but the 5 kft flight leg on transit route T1 and for all flight legs of transit route T4.

Taken together the green scattering coefficient and angstrom exponents confirm the underlying nature of the size distribution and when interpreted correctly are consistent with the results measured directly with the sizing instruments.

6.1.2 September 2003

For September the air is more polluted even on the transit routes between Adelaide and Alice Springs (T1 and T4). The scattering coefficients are above the detection threshold of the instrument for the majority of flight legs with the possible exception of the 20 kft legs.

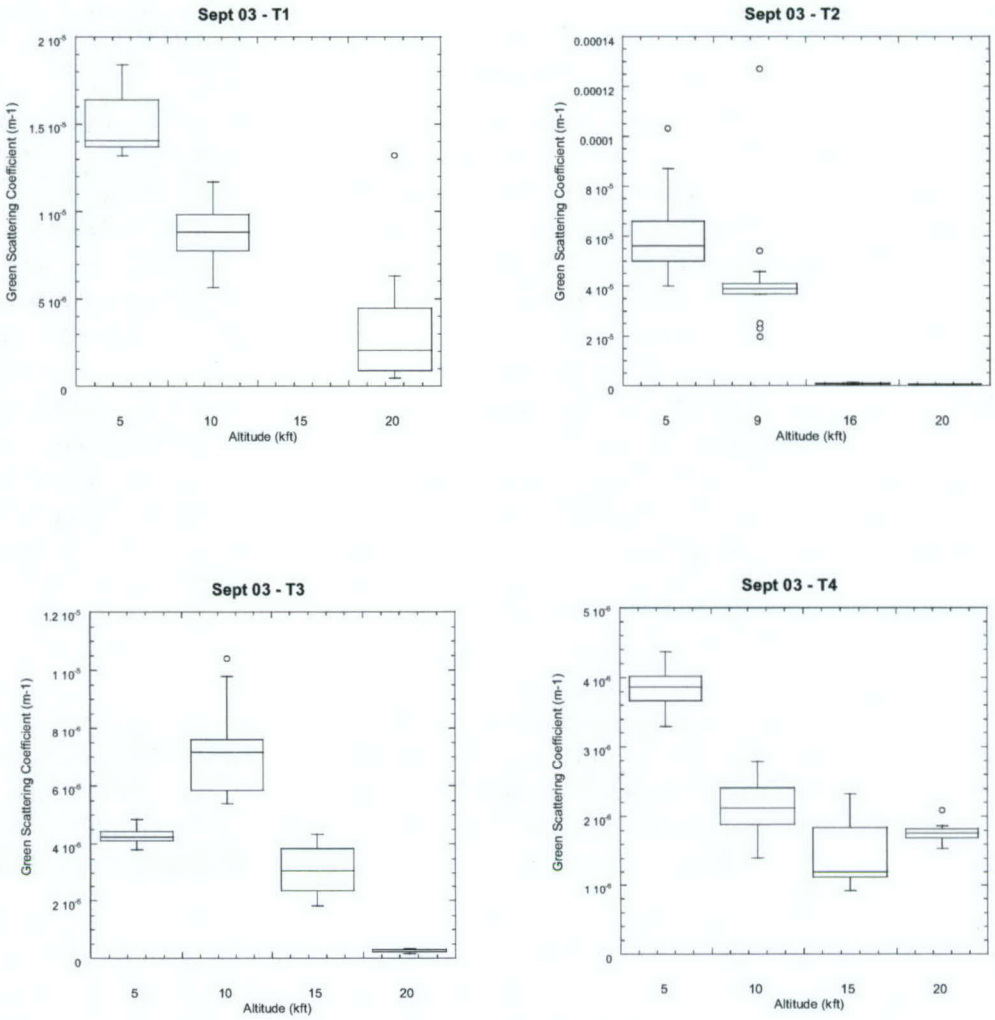

Figure 22: Modified Box plots of the green scattering coefficient for each transit route of the transit flights between Adelaide and Darwin for September 2003. Flight legs without any data are below the green wavelength detection limit except for the 15kft flight leg for transit route T1 that was not flown.

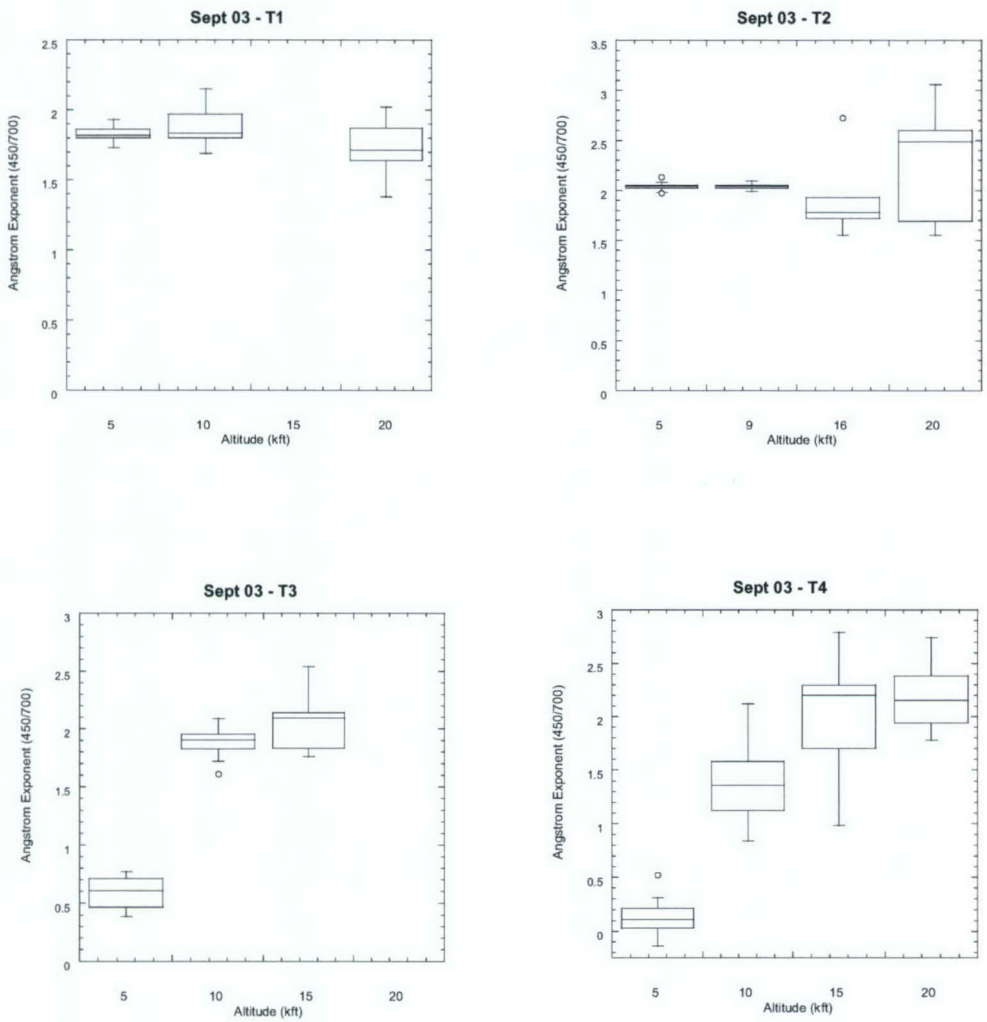

Figure 23: Modified Box plots of the Angstrom Exponent $\overset{o}{A}(450/700)$ *for each transit route of the transit flights between Adelaide and Darwin for September 2003. Flight legs without any data are below the red wavelength detection limit except for the 15kft flight leg for transit route T1 that was not flown.*

Problems with the data logging laptop during transit route T4 meant that data was being logged for only about the first half of the 20 kft flight leg and hence the results are not necessarily indicative of the aerosol prosperities along the entire leg flown. This flight leg therefore should be treated with some caution.

It is interesting to compare the Green scattering coefficient and the angstrom exponent. The constancy of $\overset{o}{A}(450/700)$ as a function of altitude indicates a balance between the amount of blue and red light scattering. This implies that the fine mode to coarse mode ratio remains constant with altitude. Even though the total amount of scattering decreases with altitude the wavelength dependence of the scattering coefficient remains approximately constant. The exception being transit routes T3 and T4 in particular.

The most striking feature of these box plots is the small values of $\overset{o}{A}(450/700)$ at lower altitudes for transit routes T3 and T4. This corresponds well with the coarse mode size distribution which shows a large peak at 5 and 10 kft and hence a significant amount of coarse particles in the atmosphere. These large particles scatter more red light than do the fine aerosols and hence the difference between the red and blue scattering coefficients becomes quite small and even negative indicating a larger red scattering coefficient. If there were also a matching increase in scattering due to the fine mode aerosol we would expect to see the angstrom coefficient remain at typical levels of 1.5 - 2. This is supported by the APS and ASASP data, which show very low levels of accumulation mode aerosol (fine mode) for these two transit routes. The large coarse mode and significantly lower accumulation mode results in the dominance of red light scattering for the lower altitudes which is reflected in the very small Angstrom exponent $\overset{o}{A}(450/700)$.

7. Conclusion and Future Work

7.1 Conclusion

These aircraft measurements of atmospheric aerosol across the centre of the Australian continent from Adelaide to Darwin are the first that we are aware of. Other measurements have been made along the East Coast and over specific regions of the country such as Katherine, Cowley Beach, Mildura and Cape Grimm.

The value of this data set is that it provides us with our first quantified analysis of transcontinental aerosol. The data in its current form has limitations due to uncertainty associated with some of the measured size distributions. In particular corrections for the unknown aerosol mass density for the APS and for the refractive index for the ASASP and FSSP are needed. As discussed in the main body of the report this data is not available. Future work may look at attempting to determine these unknown quantities. Despite this uncertainty we have confidence in the reliability of the data as the agreement in the general shape of the aerosol size distributions is quite good (Appendix A). The fact that data from a range of instruments shows this level of agreement gives us confidence that we are seeing the true underlying size distribution and not artefacts of individual sampling instruments.

In its current form this data can be used to guide the application of models of the atmospheric aerosol. We have identified differences between the Northern and Southern parts of the continent and between the months of June and September. It should be emphasised that this data was obtained on an opportunistic basis. Caution should be used in making any definitive conclusions based on these measurements alone.

The transit flight from Adelaide to Alice Springs in June produced the smallest measured aerosol number concentrations and scattering coefficients. For the majority of flight legs in June the scattering coefficients were below the detection limit of the instrument. In general the transcontinental air mass is exceedingly clean containing very little aerosol mass per unit volume.

The most notable difference between the Southern (Adelaide to Alice Springs) and Northern (Darwin to Alice Springs) transit routes was the significant impact of biomass burning aerosol North of Alice Springs. In September in particular this resulted in a very large accumulation mode peak at the lower altitudes and even the appearance of a smaller shoulder indicating the possible existence of an Aitken mode probably of fresh young smoke particles. The higher altitude (20 kft) flight leg is similar for both transit routes T1 and T2 reflecting the uniformity of the upper atmospheric chemistry and aerosol formation paths across the continent. The same is true for June with the exception being the 20kft leg for transit route T3 that shows an Aitken or nucleation mode peak, which is not present in transit route T2 for example. This is reflected in the divergence between the

total concentration and CMD for the 20kft flight leg for transit route T3 (Figure 12) whereas for the other transit routes they are closer together.

From the limited data we have for the SMPS for September the trend (shape) in the total concentration as a function of altitude for transit routes T1 and T2 remains the same for both June and September. In general the magnitude is less for June with the exception of transit route T2. This is because during June the lower altitude flight legs approaching Darwin (transit route T2) passed though a cluster of fires.

The coarse mode in June was small and we only recorded a discernible coarse mode peak for the 5 kft flight legs for transit routes T2 and T3. These levels were very low compared with September and the majority is presumably sea salt with some soil based components. This just confirms how clean the air was during June other then when passing close to or directly through biomass fires. This can be contrasted with September where significant coarse mode size distributions were measured for all four-transit routes at both 5 and 10 kft. The most noticeable difference in September between transit routes T1 and T2 was the increase in concentration below 1 μm for transit route T2 due to the large amount of smoke in the atmosphere. There is a similar increase in the concentration below 1 μm for transit route T1 due to some smoke being measured from the bushfires on the East Coast of Australia (Figure 6). Smoke in this case is transported by the low altitude winds out over the Pacific ocean then back across the north of Australia circulating down through the centre and then back to the East coast (Figure 2). The APS measures the very tail of the accumulation mode (due to smoke), which is measured by the SMPS. This can be contrasted with transit routes T3 and T4, which show a sharp drop below 1 μm. This appears to be a result of the winds being anomalous in behaviour (Figure 3) and hence none of the East coast bushfire smoke is transported into the flight paths. As a result the accumulation mode is very small. This is also confirmed by the ASASP and nephelometer data for these transit routes. Besides the noticeable sharp drop in concentration below 1 μm we also observe a small shoulder at about 1.5 μm for the 5 kft leg for transit routes T3 and T4 (Figure 19). It would be nice to have access to some chemistry data for these transit routes but we did not operate a low altitude filter on the return transit from Darwin to Adelaide. It perhaps is significant that the continental air mass has altered significantly within eight days. It also needs to be kept in mind that the lower and higher altitude flight legs are not over the same locations on the ground between the outbound (T1 & T2) and inbound transits (T2 & T3). At this stage the small shoulder remains unexplained.

The majority of the coarse mode in September is likely to be sea salt however a small amount of intercontinental dust or smoke can not be ruled out based on the TOMS data for September.

The scattering coefficients for September reinforce this, particularly the lower altitude values for transit routes T3 and T4, where the angstrom exponent is very small around 0.5 (and some values even negative for transit route T4) indicating the dominance of large particles, which scatter a greater amount of red light. Generally the Green scattering coefficient decreases with increasing altitude before flattening off. The exception being the

10 kft flight leg of transit route T3. Unfortunately we do not have any SMPS data for this transit route so we are unable to correlate this with the fine mode concentration. However it correlates strongly with the coarse mode concentration (Figure 18) or more accurately the APS concentration that includes the tail of the accumulation mode. The 10 kft flight leg passed very close to a small group of fires to the West (Figure 6) before the descent to 5 kft and hence this would have resulted in a large increase in the accumulation mode concentration. Thus at 10kft the CMD is lower than at 5kft (finer smoke particles) but the number concentration increases due to the fires (Figure 18). These fires have appeared to have little impact on the 5 kft flight leg. The median angstrom exponent is generally around 2 other than the previously mentioned 5kft flight legs for transit routes T3 and T4. The value of 1.5 for the 10 kft flight leg for transit route T4 indicates that there was a reduction in the fine mode aerosol and hence less scattering at a blue wavelength compared with red. The volume distributions confirm this indirectly since the coarse mode for the 10 kft flight leg for transit route T4 is actually smaller than for transit route T3 so it cannot be an increase in coarse mode particles producing the reduction in the angstrom exponent therefore it must be a decrease in fine mode particles resulting in the angstrom exponent decreasing from the median value of around 2. June provides very little useful information on the variation in the scattering properties of the aerosol other than to confirm that the air was exceedingly clean resulting in very little scattering other then for the transit flight into Darwin that passed through the cluster of fires just South of Darwin (Figures 4 & 20).

It needs to be emphasized that this data is a snapshot in time of the microphysics and optical properties of the transcontinental aerosol. The influence of local aerosol sources and continental weather patterns have been highlighted. The features that are most easy to correlate with the data are the biomass fires and the horizontal wind direction. To make any statistically significant conclusion about the continental aerosol microphysics and optical properties a larger number of measurements would be necessary. However these measurements were opportunistic in nature and at the very least provide some quantitative data on how the aerosols can change between months (time) and between the Southern and Northern parts of the continent (space).

Finally in Appendix A we present all the combined size distribution data measured during the transit flights. We have chosen to keep all individual flight leg size distributions as there was insufficient data to perform any useful averaging other than over individual flight leg samples. This represents a different philosophy between the analysis of these transcontinental measurements and the measurement campaigns that were conducted over Jabiru during June and September 2003 [1]. In the latter case we collected many more samples at a better vertical resolution and over longer horizontal flight paths. We also made morning and afternoon flights and flight legs were always over the exact same region on the ground. This improved our counting statistics and provided for a more representative analysis of the difference between the aerosol microphysics, chemistry and optical properties over Jabiru between June and September 2003.

For the transcontinental flights too few samples were measured and hence rather than perform averaging, which would obscure differences between flight legs, we have chosen to show each flight leg separately for the combined size distribution data.

7.2 Future Work

In the absence of more detailed chemistry data an attempt could be made to retrieve particle effective density and refractive index using the method (technique) proposed by Hand and Kreidenweis [23]. However this would be extremely challenging using the aircraft data where the concentration is very low and because there is not enough samples to build up the necessary confidence in the particle counts for a given size bin, particularly at larger sizes. This will be an issue in the overlap regions between the data where a best fit is made by combining the data in a statistically rigorous manner. It would also be challenging combining data from so many instruments all with their own corrections (other then for the SMPS). As far as we know this method has only been applied to good quality data obtained from ground-based measurements.

From a scientific point of view it would be more interesting to combine the analysis from the aircraft measurements presented in this report with long term ground based data obtained from CSIRO's network of sun photometers of the atmospheric aerosol optical depth (AOD). Comparison of AOD calculated directly from the in situ instruments on the aircraft and those obtained from the sun photometers would allow a cross check between both methods. The sun photometer data is obtained on a continuous basis and hence we can use it to monitor the long-term trend in the transcontinental aerosol. The aircraft data is a snapshot in time but provides for some detail on the vertical resolution. The two approaches are complementary.

Additionally comparisons can also be made with TOMS AI data to see if there exists consistency between all three data sets. This type of analysis would be better suited to regions of relatively high aerosol concentration such as the top end of the Northern Territory.

8. Acknowledgements

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The efforts of the CSIRO team lead by Dr John Grass and including Mr Bernard Petraitis and Mr Charles Tivendale are appreciated. Finally thanks are given to Dr Zoran Ristovski from QUT who was responsible for the installation, testing and data collection for the QUT SMPS during the transits flights in June and to Dr Milan Jamriska for the same in September. The technical support of Mr Mark Burridge during the installation and testing of DSTO equipment and for technical support during the deployment is also acknowledged. Thanks are also given to Ms Michelle Hackett for assistance with some of the data analysis.

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Appendix A: Combined Differential Size Distribution Plots

The quality of the data is better in September. This is primarily due to the increased concentration of aerosols in the atmosphere. The ASASP performance was also improved during September.

The APS counting efficiency is not uniform across the entire size range of the instrument [1]. Below 0.7 μm the counting losses can be high (50% at 5 μm) dropping to around 0% at 1 μm and then increasing again at 5 μm. When both probes are operating correctly we prefer to use the concentration from the ASASP below 0.7 μm and from the FSSP above 5 μm. These are not absolute cut-offs but are typical values for the diameters at which we expect the APS to suffer major particle loss. This is especially the case for particle sizes larger than 5 μm due to losses in the isokinetic inlet. Some of the APS data show this effect especially at smaller sizes showing a drop in particle concentration. This is an artefact of the instrument and is not representative of the shape of the underlying size distribution. This is borne out when comparing with the ASASP data, which generally is increasing with decreasing particle size as it is measuring the tail end of the increasing accumulation mode.

At larger particle sizes the concentration in the atmosphere is very low which is reflected in the APS data with the tail becoming flat (at or below the 0.001 (cm^{-3}) detection limit of the instrument) or exhibiting large variation (count noise).

The SMPS exhibits noise at larger particle sizes, which is as a result of lower particle concentration but also a sampling artefact of the instrument. It is being operated in fast scan mode and as a result smearing of the tail of the SMPS distribution can occur. For smaller particle sizes the data can also become noisy due to the sampling limitation of the SMPS on an aircraft. It takes time for the instrument to stabilize with respect to pressure at a new altitude so some of the data may reflect this lack of stability. We note also that the SMPS did not perform well on the return flight legs from Darwin to Adelaide during September (transit routes T3 and T4). Therefore there was no useful data obtained for the instrument on these flights legs and we have to rely on the ASASP and APS data.

The FSSP in June for all but the lowest altitude is zero in all but one sizing channel (the lowest). This just reflects how clean the continental air mass was particularly with very little coarse mode material aloft. The exception is the 5kft and 10 kft flight legs for transit route T4 when the aircraft passed through cloud and the FSSP and the ASASP recorded large particle (cloud droplet) counts.

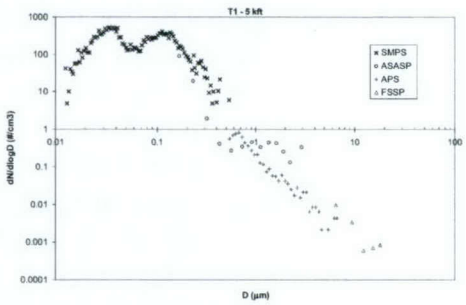
We have chosen to display the data from all instruments across the entire size range measured. This includes data that is likely to be noise. However in a majority of cases the

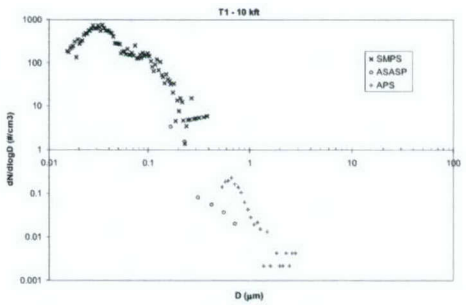
shape of the distribution can be observed. In the next two sections we make some comments on individual flight legs.

These measurements should be very much viewed as a snapshot in time and they reflect real differences in the aerosol properties across the continent for the particular times of the year. In no way do we claim to have summarised all possible conditions that may be encountered on any transcontinental crossing. It is possible that these results will have changed significantly a week earlier or later then when they were conducted. However the gross atmospheric phenomena such as wind speed and direction were observed to be consistent with historical met data. The sources of aerosol are always going to come from biomass fires up North, sea salt especially close to Adelaide and Darwin, some soil based or desert dust over the arid and semi arid interiors of the country and from long range transport of inter-continental aerosol that may include additional amounts of smoke and dust. Aerosol formation mechanisms in the FT are fairly universal but will be affected by the presence of biomass fires and anthropogenic sources from major urban centres. Whilst the relative amounts of aerosol species may vary day-to-day or week-to-week the overall results should reliably summarize how the aerosol properties change across the continent under standard atmospheric conditions. In the event of a major event such as El Nino or La Nina one could expect differences to be observed. This is where long term seasonal aerosol monitoring becomes important so that we are able to start to build up a continental aerosol climatology for Australia. The aircraft data then becomes important as a means of validating some of the assumption made about how the aerosol changes with altitude that are used to determine column integrated quantities such as AOD using ground based sun photometers positioned around the continent.

One final comment about these plots is that they are on a log-log scale due to the fact that they span four orders of magnitude in size and up to eight orders of magnitude in number concentration. In some cases this makes the data appear better than they actually are and also sometimes worse than they are. The APS and FSSP concentrations being so low are suited to a log plot generally because they are less than 1 (cm^{-3}). The SMPS is suited to a linear plot with the concentration being as high as 1000 (cm^{-3}).

A.1. June 2003


T1 - 5 kft
SMPS
ASASP
APS
FSSP
dN/dlogD (#/cm3)
D (µm)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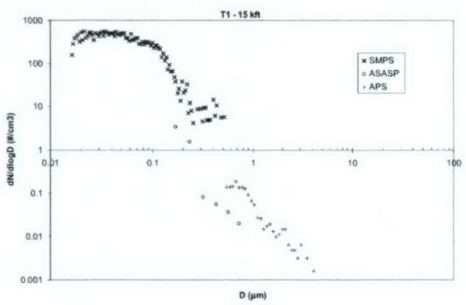
SMPS
ASASP
APS
dN/dlogD (#/cm3)
D (µm)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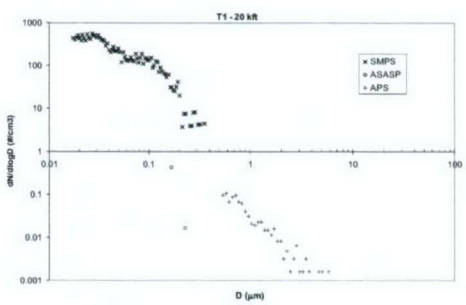
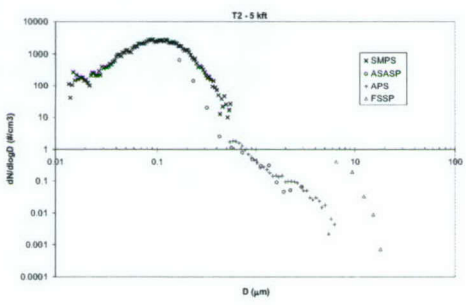
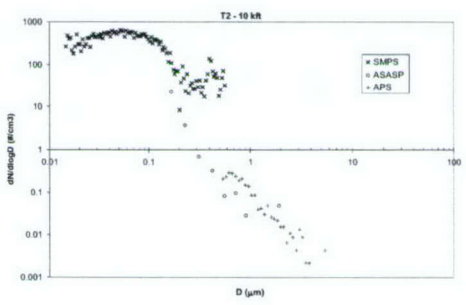
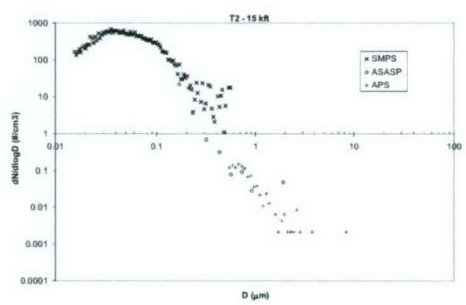
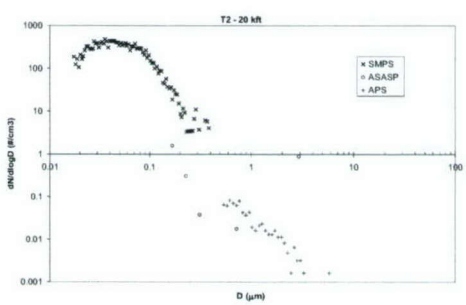
SMPS
ASASP
APS
dN/dlogD (#/cm3)
D (µm)


T1 - 20 kft
SMPS
ASASP
APS
dN/dlogD (#/cm3)
D (µm)


T2 - 5 kft
SMPS
ASASP
APS
FSSP
dN/dlogD (#/cm3)
D (µm)


T2 - 10 kft
SMPS
ASASP
APS
dN/dlogD (#/cm3)
D (µm)


T2 - 15 kft
SMPS
ASASP
APS
dN/dlogD (#/cm3)
D (µm)


T2 - 20 kft
SMPS
ASASP
APS
dN/dlogD (#/cm3)
D (µm)

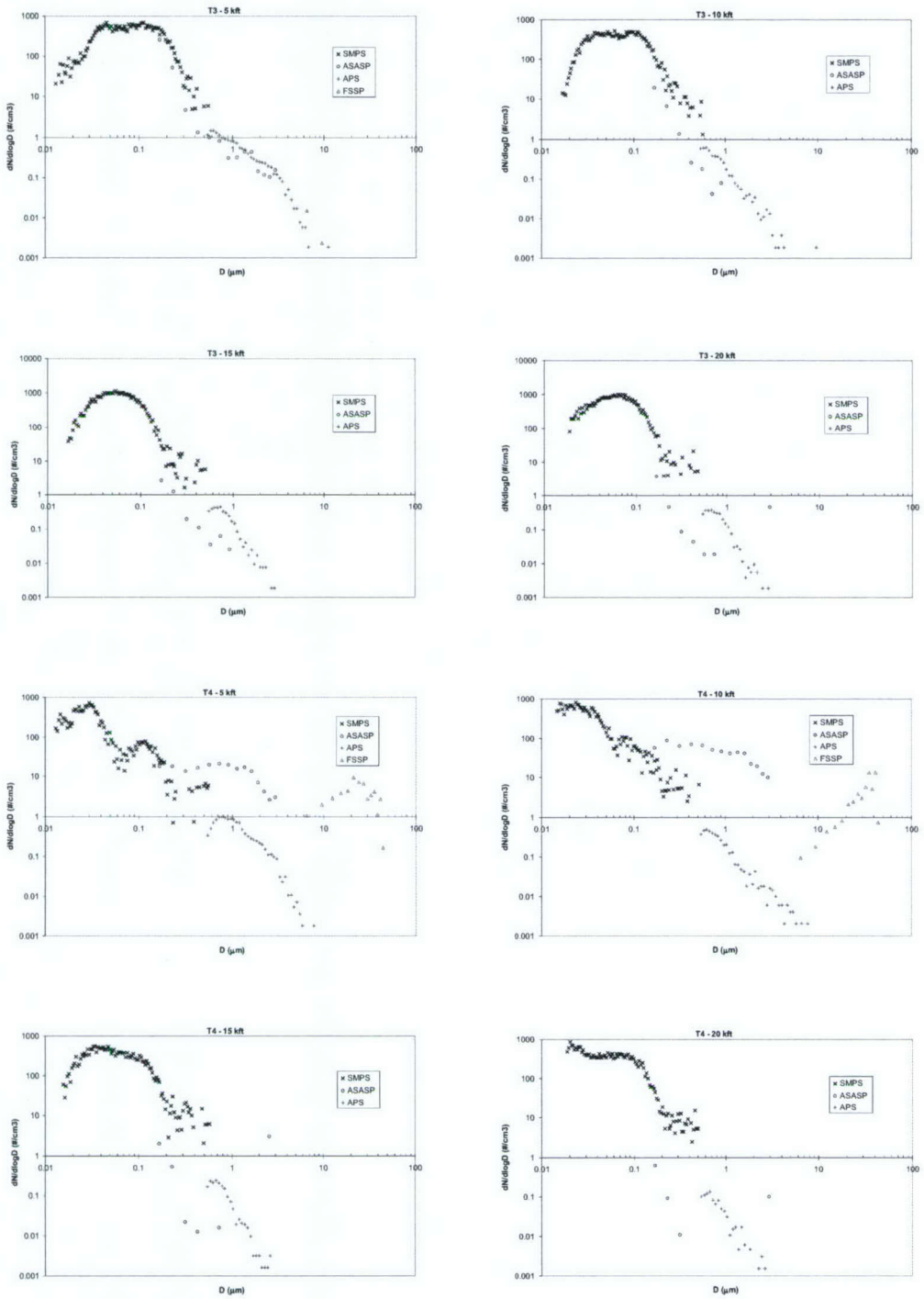


Figure 24: Combined size distribution plots for all four sizing instruments (when available). There are total of 16 transit flights in June spread over four transit routes and four different altitudes for each transit route.

We have left out the smallest size bin (channel) for the ASASP and the FSSP, as it is likely to be noise due to the laser detuning. This applies for both the June and September data. A number of the SMPS distributions exhibit noise towards the tail generally above about 0.2 μm. This data should not be considered reliable. There is large disagreement between the ASASP and the APS in many cases particularly at higher altitudes (10 and 15 kft). We know the ASASP was not operating optimally during June but the APS was performing within operating limits and hence is consistent across all flights for June and September.

In the majority of cases the FSSP data is zero for the higher altitudes for all but the lowest size channel, which we have left out in any case. The real anomaly is transit route T2 5kft flight leg with the FSSP exhibiting a very large concentration of particles above 3 microns that is not observed in the APS data or the ASASP data. This could genuinely be an increase in these larger size particles or something erroneous with the instrument. This flight leg corresponds to the approach to Darwin through a cluster of fires just south of Darwin. Potentially it may include large bits of debris emanating from the fires (fly ash) though it is not clear this material would be transported to 5 kft. Certainly the magnitude of the difference between the APS and FSSP for this flight leg is much larger than the norm indicating it is more than just particle losses through the isokinetic inlet that is occurring. This is also supported by the fact the ASASP counts remain low in the larger size bins. This just goes to highlight how important it is to have multiple sizing instruments that measure the size of particles using different techniques and also cover separate size ranges with some overlap.

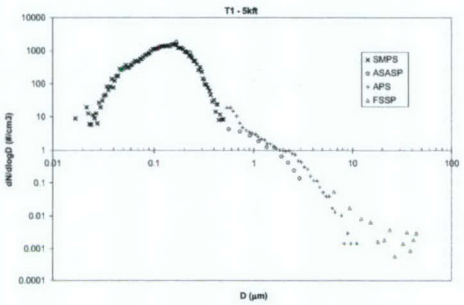
There are marked differences between the ASASP and APS for most of the 15 and 20 kft flight legs. The exception being the 15 kft flight leg of transit route T2. In these cases the APS is consistent with the SMPS and in fact one could extend where the SMPS finishes off to where the APS commences basically with a straight line. The ASASP has been shown for completeness but certainly the SMPS and the APS should be considered more reliable based on these distributions.

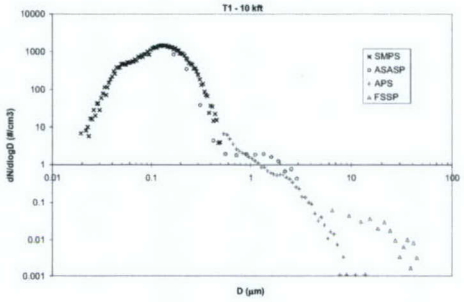
The apparently unusual distributions for transit route T4 5 and 10 kft flight legs are a result of the aircraft passing through cloud. This shows up in the SMPS and APS distributions with slight modifications of the shape but is pronounced in the probes, which measure the clouds in situ. The FSSP is actually a cloud probe and is designed to measure the changes in cloud properties at a very high temporal resolution. Even in these cases there is still an offset observed between the ASASP and FSSP distributions for the 10 kft flight leg.

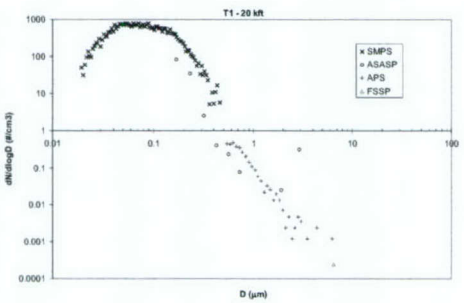
Generally speaking the accumulation and coarse mode aerosol concentrations where very low. The amount of mass collected on the filters was exceedingly low. This just highlights how clean the continental air is over Australia. The major source of accumulation mode aerosol was biomass smoke for the Northern transit routes as expected. The coarse mode was also only significant for the lower altitudes of the Northern transit routes and is most likely due to sea salt. This probably makes up the coarse mode for the Southern transit

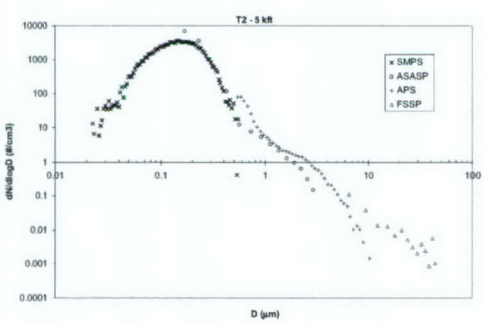
routes as well but at a low concentration such that there is no discernible peak in the volume distributions (section 5.2.1).

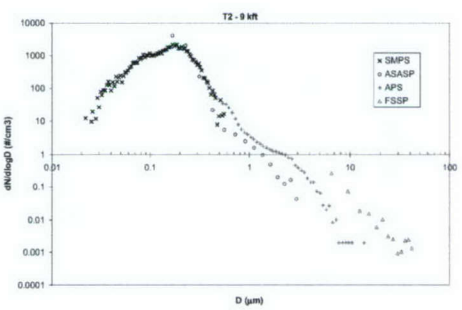
A.2. September 2003

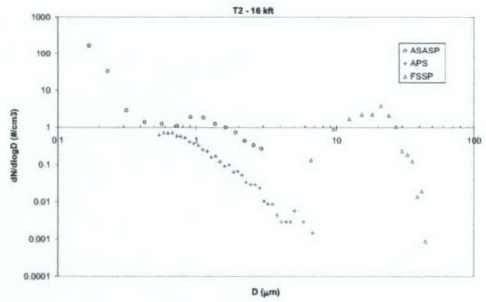


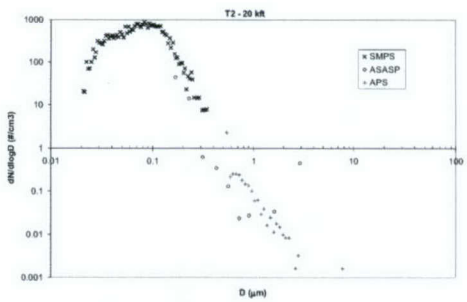


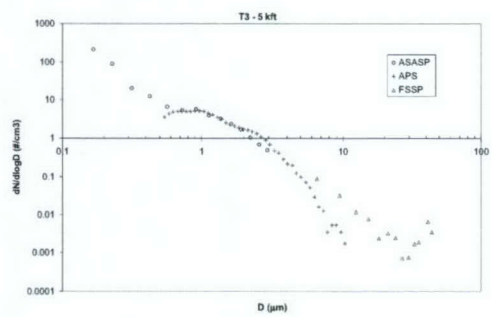


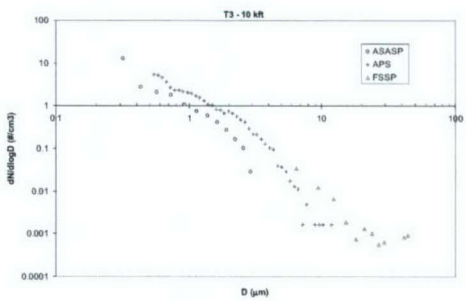


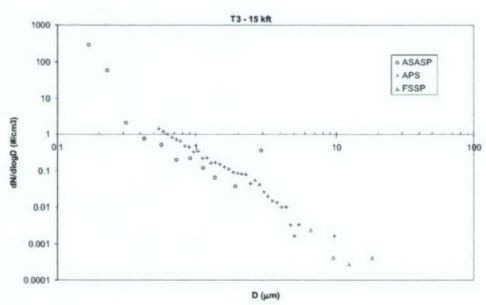


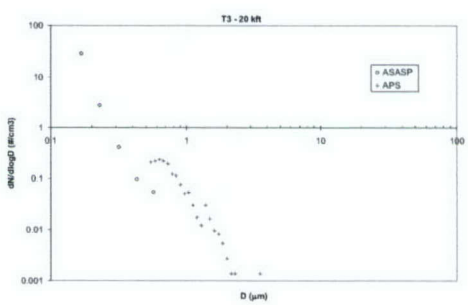

T2 - 16 kft
ASASP
APS
FSSP
dN/dlogD (#/cm3)
D (μm)

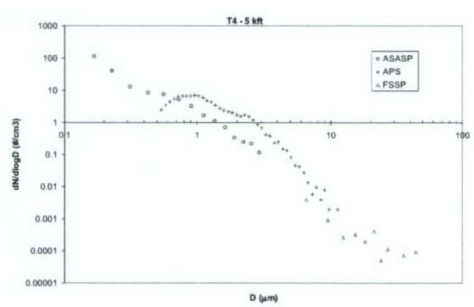

T2 - 20 kft
SMPS
ASASP
APS
dN/dlogD (#/cm3)
D (μm)

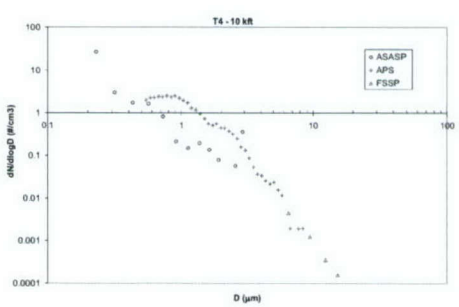

T3 - 5 kft
ASASP
APS
FSSP
dN/dlogD (#/cm3)
D (μm)


T3 - 10 kft
ASASP
APS
FSSP
dN/dlogD (#/cm3)
D (μm)


T3 - 15 kft
ASASP
APS
FSSP
dN/dlogD (#/cm3)
D (μm)


T3 - 20 kft
ASASP
APS
dN/dlogD (#/cm3)
D (μm)


T4 - 5 kft
ASASP
APS
FSSP
dN/dlogD (#/cm3)
D (μm)


T4 - 10 kft
ASASP
APS
FSSP
dN/dlogD (#/cm3)
D (μm)

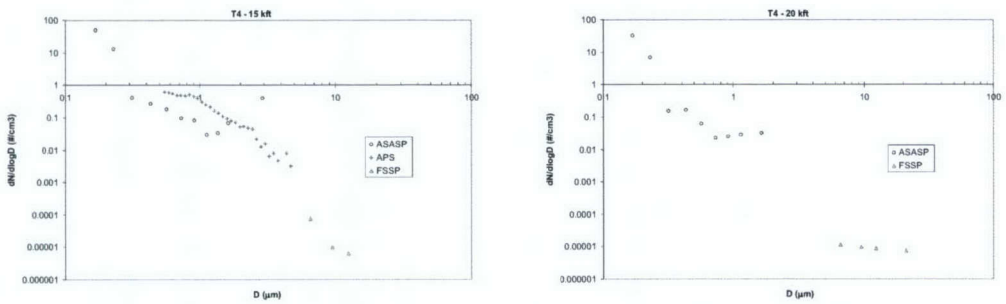


Figure 25: Combined size distribution plots for all four sizing instruments (when available). There are total of 15 transit flights in September spread over four transit routes and four different altitudes for each transit route (when flown).

Concentration levels are much higher in September for fine and coarse mode aerosols. Unfortunately we did not collect any useful data from the SMPS for transit routes T3 and T4. The APS data logging laptop experienced problems on the 20 kft flight leg of transit route T4 so this data is also missing. In the case of the return transits (T3 and T4) we principally rely on the ASASP to provide some information about the accumulation mode aerosol.

Generally there is much better agreement between all four sizing instruments during September particularly in terms of shape and also concentration. The APS does a much better job of determining the coarse mode peak then was the case for June. The FSSP data is also much more useful and does a better job of providing the coarse mode tail as expected since the concentration of coarse mode aerosol beyond 10 µm was at measurable levels for all altitudes with the exception of most of the 20 kft flight legs. The SMPS tails are much better behaved for September with the exception of some of the smallest size intervals for a few flight legs.

Whilst there are still differences between the ASASP and APS they are less pronounced then for June and where there is a difference it is mainly in the concentration with both instruments doing a good job of determining the shape of the coarse mode aerosol distribution. For some of the higher altitude legs the disagreement is most pronounced. As discussed earlier (section 5.4) we would expect agreement to improve if corrections were applied to the size distribution data.

The interesting flight legs are the 5 and 10 kft for transit route T4. If we examine the volume distribution for the APS (section 5.2.2) we see that there is small shoulder around 1 µm and the main coarse mode peak as expected around 3 µm. This additional structure also appears in the ASASP data, which reinforces our belief that this is a real result. However compared with the transit route T3 flight leg at the same altitude the ASASP seems to underestimate the APS concentration quite significantly. This was the highest

coarse mode concentration recorded on any of the transit routes. The difference may reflect how significant the density correction will be for the APS and the refractive index correction for the ASASP for these particular aerosol and flight leg conditions. It is also worth noting the good agreement between the tail of the APS and the FSSP data for these two flight legs compared with the same two flight legs for transit route T3. In the latter case the APS under estimates the tail compared with the FSSP. For transit route T4 the FSSP joins smoothly onto the tail of the APS. This indicates that not too much particle loss occurred through the isokinetic inlet since presumably the large particle number concentration was small to begin with. This is why the FSSP concentration above 5 μm is small (compare with transit route T3). This tells us that the coarse mode for these two flight legs was dominated by particles with a sharper cut-off in size then for the majority of other flight legs that recorded a substantial coarse mode concentration. It would be interesting to know more about the chemical composition of the T3 and T4 coarse modes both for a relative comparison and also to account for the unusual shape of the coarse mode volume distributions for transit route T4.

The large increase in the FSSP distribution for the transit route T2 16 kft flight leg is most likely an indication that the aircraft passed through some cloud. This would also account for why the ASASP concentration is much higher than the APS concentration for this particular flight leg. The FSSP distribution tends to sit above the APS distribution as expected and provides a more realistic estimate of the tail of the coarse mode size distribution. The difference is most pronounced for the 9 kft flight leg of transit route T2. Based on the shape of the FSSP distribution for these flight legs this is not likely to be cloud since the ASASP distribution looks like a normal aerosol size distribution. One should bear in mind that the coarse mode concentration particularly for the FSSP is exceedingly low with very little aerosol mass suspended in the air.

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S. B. Carr

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19. ABSTRACT

Aircraft measurements of atmospheric aerosol were made during a number of transcontinental flights across Australia from Adelaide to Darwin during June and September 2003. A range of aerosol measuring equipment was operated during these flights enabling data to be collected on the aerosol chemistry, physical size distribution and scattering coefficient. Additional information has been obtained from other sources on the continental weather patterns and global atmospheric aerosol loading for the period that covers these flights. Together this information presents a picture of the aerosol sources, microchemistry, microphysics and optical properties allowing for differences to be explored between Southern and Northern regions of Australia at different altitudes (spatial information) and between the months of June and September (temporal information). The conclusions are that the aerosol mass density is small in general particularly during the month of June. At lower altitudes smoke tends to be the dominant accumulation mode aerosol and sea salt a major constituent of the coarse mode. There is evidence of some influence of intercontinental aerosol during September but the level of importance is difficult to quantify.

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