COVER SHEET

Trends in size classified particle number concentration in subtropical Brisbane, Australia, based on a five year study

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Abstract

Particle number size distribution data in the range from 0.015 to 0.630 μm were collected over a five-year period in the central business district (CBD) of Brisbane, Australia. Particle size distribution was summarised by total number concentration and number median diameter (NMD) as well as the number concentration of the 0.015-0.030 (N_{15-30}), 0.030-0.050 (N_{30-50}), 0.050-0.100 (N_{50-100}), 0.100-0.300 ($N_{100-300}$) and 0.300-0.630 ($N_{300-630}$) μm size classes. Morning (6:00-10:00) and afternoon (16:00-19:00) measurements, the former representing fresh traffic emissions (based on the local meteorological conditions) and the latter well-mixed emissions from the CBD, during weekdays were extracted and the respective monthly mean values were estimated for time series analysis. For all size fractions, average morning concentrations were about 1.5 higher than in the afternoon whereas NMD did not vary between the morning and afternoon. The trend and seasonal components were extracted through weighted linear regression models, using the monthly variance as weights. Only the morning measurements exhibited significant trends. During this time of the day, total particle number increased by 105.7% and the increase was

greater for larger particles, resulting in a shift in NMD by 7.9%. Although no seasonal component was detected the evidence against it remained weak due to the limitations of the database.

Keywords: Submicrometre particles, ultrafine particles, number size distribution, time series, regression models.

1. Introduction

Until recently, ambient airborne particulate matter monitoring has focused on its mass concentration, usually as PM_{10} (mass concentration of particles < 10.0 μm) and $PM_{2.5}$ (≤ 2.5 μm). Over the last ten years or so, with growing concerns about the potential health effects of smaller size fractions, an increasing number of studies began to measure submicrometre (< 1.0 μm) and ultrafine particles (< 0.1 μm). Although their contribution to the total particle mass is very low, in urban air they dominate the particle number (eg. Jaenicke, 1993) with over 80% of the total count lying in the ultrafine range (Morawska *et al.*, 1998a; 1998b). Number concentrations above 10^4 cm^{-3} are common in the literature, and some studies have reported concentrations above 10^5 cm^{-3}(Hussein *et al.*, 2004; Shi *et al.*, 1999). However, since at present there are no air quality standards for particle number concentration, routine monitoring of particle number has not been conducted and therefore long-term trends of this important particle characteristics are unknown. Yet, this information is required in order to investigate any epidemiological associations.

Some insight into particle number concentration trends was provided by the studies conducted at different locations in former East Germany, including the city of Erfurt and the counties of Bitterfeld, Hettstedt and Zerbst, during two different campaigns, the first one in the early and the second one in the late 1990's, which found that

ultrafine particle number increased between 38.1% (Pitz *et al.*, 2001) and 115% (Ebelt *et al.*, 2001). Unfortunately, most measurement campaigns have been of very short duration, lasting only a few months (Buzorius *et al.*, 1999; Wahlin *et al.*, 2001, and others). Only a handful of investigations have continuously monitored submicrometre particles for periods of at least one year: these include a report of different campaigns lasting between one and three years in five European cities (Paatero *et al.*, 2005), and reports of 13-months monitoring in the urban area of Atlanta (Woo *et al.*, 2001), 12 months at supersites in Pittsburg, Philadelphia (Cabada *et al.*, 2004) and Fresno, California (Watson *et al.*, 2005), four years in Leipzig, Germany (Wehner and Wiedensohler, 2003) and two years in the subtropical atmosphere of Brisbane, Australia (Morawska *et al.*, 1998b). Findings from these studies include strong particle number associations with nitrogen oxides (Morawska *et al.*, 1998b; Paatero *et al.*, 2005), correlations with global radiation levels and wind conditions (Wehner and Wiedensohler, 2003); seasonal differences in chemical composition and higher number concentration in winter (Cabada *et al.*, 2004), and low particle concentrations associations with low levels of sulphur precursors and relative humidity (Watson *et al.*, 2005), but no evidence on the long-term trends. The only exception is a Helsinki study by Hussein *et al.* (2004), who measured particle number between May 1997 and February 2003, and found that annual geometric mean particle number concentration increased by 3.2% in 1999, followed by a decreased of 6.7% in 2000 and 17.6% in 2002; although the monitoring site was moved after the first three years 3km from its original location, thereby influencing the results, their main conclusion was that the annual variation in total particle number was associated with traffic density and the predominance of new vehicles in the Helsinki area.

To address this limitation in knowledge this paper presents the results of time series analysis of size-fractionated particles number concentrations in the ranges 0.015-0.030, 0.030-0.050, 0.50-0.100, 0.100-0.300 and 0.300-0.630 μm, as well as the total in the range from 0.015-0.630 μm, collected over a five-year period in the subtropical city of Brisbane, Australia. The objective was to identify the short and long-term components and test the hypothesis that submicrometre particle number has increased over time. This hypothesis was formulated based on the evidence of the increase in vehicle traffic, associated with population growth and increased vehicle usage, in the South East Queensland, where Brisbane is located (Neale and Wainwright, 2001b).

2. Methods

2.1. The city of Brisbane

Brisbane, the capital city of the state of Queensland, Australia, is situated at $27^{o}\,28^{'}$ latitude and $153^{o}\,02^{'}$ longitude. The greater Brisbane region lies in the coastal plains east of the Great Dividing Range, although the urban area is dotted by large hills reaching up to 300 metres. Generally, the city is a low-lying floodplain, presenting a potential for blocking and channeling of local winds, particularly in the upper valley areas.

Brisbane has a subtropical climate, typical of the eastern coastal locations of the high pressure belt of the southern hemisphere (Simpson, 1989). In general, during the warm season (November-April), unstable sea breeze flows from the Pacific Ocean; the weather is dominated by strong winds with average speeds of 11-21 km h^{-1} and frequent strong rain. During the cool half of the year (May-October), the large scale of air drift is from the southwest quadrant and this continental air is relatively dry (0-

50% relative humidity) and stable, low winds speeds prevail and pollutant persistence times are increased in the absence of precipitation.

Its population enjoys a high level of vehicle ownership with private and commercial vehicles being the dominant mode of transport. Over the past two decades, the city has experienced the fastest population explosion in the country and therefore a substantial increase in vehicle traffic.

2.2. Description of the monitoring site

The data used for this analysis was collected at the Air Monitoring and Research Station (AMRS) of the Environmental Protection Agency (EPA), situated in the sixth floor of a building at the Queensland University of Technology (QUT). The project was a joint venture between the university and the EPA. Campus is located at the south-western end of the central business district (CBD) and is bounded by a busy freeway 100 m southeast to the station, and the City Botanic Gardens in the opposite side (Figure1)

The CBD covers an area of approximately 2.2 km^2 and is located in a curve of the Brisbane River. A general meteorological trend around the area is the reversal of wind direction between the morning (wind originating from the SE quadrant towards the city) and the afternoon (wind moving towards the SE), owing to the opposite directions of sea and land breezes. This means that in the morning the wind moves predominantly from the freeway towards the station and in the afternoon it moves in the opposite direction, bringing to the station well-mixed air from the CBD.

By far, traffic is the principal pollution source around the CBD. There are also a limited number of small industries in the nearby suburbs including a brewery, and further away, a power station, oil refineries and an oil terminal. Biomass burning is a common land management practice in the surrounding forestry and agricultural areas

during the low wind speed period of July to September. On these occasions, the city may be covered in smoke channeled through the drainage flows.

2.3. Sampling procedure

Particle number size distribution (NSD) was measured using a TSI Model 3394 scanning mobility particle sizer (SMPS) covering the size range 0.015-0.630 μm (referred in this study as submicrometre particles). The details of the operation of the instrument have been provided in an earlier study (Morawska *et al.*, 1998b). Monitoring was conducted between July 1995 and November 2000. Prior to 1998, the SMPS was used for regular "grab sampling" of NSD in triplicates everyday in the mornings (9:30 – 10.00) and afternoons (16:30 – 17.00). On days characterised by notable atmospheric or meteorological conditions such as haziness or hazard reduction burning, NSD were measured at regular intervals during the whole day. Since January 1998, the instrument was used to collect data at hourly intervals on a continuous basis. The scan time per sample was 90s seconds, and the data were averaged over 1-hour intervals. A total of 9,060 datapoints were obtained, the earliest corresponding to 10 July 1995 and the last one to 7 November 2000.

During this entire period also concentrations of other pollutants were monitored at the station, including PM_{10}, NO_x, SO_2, CO, as well as light scattering. Analysis of the correlation between concentration of these pollutants based on the first two years of monitoring was presented earlier (Morawska *et al.*, 1998b).

2.4. Description of the database and calculation of selected particle parameters

Particle number size distribution (NSD) data was grouped into 64 size channels. The number median diameter (NMD) of the distribution was calculated using the formula:

$$NMD = L + \left(\frac{\frac{N_{Total}}{2} - N_{cum}}{N_{Median}} \right) w \qquad \textbf{(1)}$$

where L is the lower limit of the channel containing the NMD, N_{Total} is the total particle number concentration, N_{cum} the cumulative concentration of the channels preceding the one containing the NMD, N_{median} and w the number concentration and the width, respectively, of the channel containing the NMD.

The data was then divided into different size fractions in order to analyse and compare their behaviour over time. The following size classes were used in this analysis: 0.015-0.030 (N_{15-30}), 0.030-0.050 (N_{30-50}), 0.050-0.100 (N_{50-100}), 0.100-0.300 ($N_{100-300}$) and 0.300-0.630 ($N_{300-630}$) μm. The number concentration (N_{p-q}) of each size class (p-q in nm) was calculated by adding the concentrations of all the channels contained within that class. In other words:

$$N_{p-q} = \alpha \sum_{p}^{q} N(d_p) \qquad \textbf{(2)}$$

Where $N(d_p)$ is the number concentration of the channel covering the size interval from d_p to $d_p + \Delta d_p$. The lower and upper limits of each size channel had to be calculated from the midpoint diameter. Therefore, it was necessary to introduce a correction factor α, which was obtained from the equation:

$$\alpha = \log_{10}(d_p + \Delta d_p) - \log_{10}(d_p) \qquad \textbf{(3)}$$

Calculations of the log difference between consecutive channels gave an average α-value of 0.03125 and this was the value used in equation (2).

2.5. Data analysis methods

2.5.1 Daily variations in particle number

The database was divided into hourly intervals and the measurements were averaged over each interval. Time of the day was defined as the grouping variable and the hypothesis that particle number varied with time of the day was tested through one-way analysis of variance (ANOVA). The procedure was done separately for each size class. Homogeneity of variance between groups, or homocedasticity, was tested using Levene's statistics. The F-ratio was used when this condition was satisfied and Brown-Forsythe statistics when it was violated.

Post hoc tests aimed to gain further insight into the association between time of the day and particle concentration levels. The aim was to identify data subsets that could explain the underlying dynamics and their variations throughout the day. Each pair of means was compared in order to determine whether the differences were statistically significant. Tukey tests were applied in the case of equal variances and Games-Howell tests when they were unequal. Both ANOVA and post hoc tests were computed with SPSS (version 12.0.1, SPSS Inc., 2003).

2.5.2 Outliers and missing data

The objective of analysing the quality and level of completeness of the database was to identify sources of estimate bias that could lead to incorrect interpretations of the data. Outliers and missing observations were expected in the present investigation and therefore their influence on the estimates was evaluated. Some outliers were predicted to be kept as valid observations and others to be eliminated thereby increasing the amount of missing information.

In the present dataset, outliers had to be defined and carefully scrutinised before their retention or elimination. The aim was to minimise their influence on the estimates and ensure the retention of legitimate measurements. The first step was to define upper and lower total particle number concentration limits based on the expected concentration range in urban air. The minimum concentration reported for Brisbane was 4.0 x 10^2 cm^{-3} (Morawska *et al.*, 1998b). In this study, the lower limit was set to 1.0 cm^{-3} with the values around this limit anticipated to be quite rare. The upper limit was set to 2.0 x 10^5 cm^{-3}. Higher concentrations have been found elsewhere, for example during a 4-day study in Birmingham (Shi *et al.*, 1999), but they were rare in the present database. The number size distributions around and outside these limits were then analysed using robust statistical techniques, including the analysis of local or temporal correlations with the other pollutants measured at the station. 46 datapoints (around 0.5 percent of all the available data), considered to be outliers, were removed. Of these, 9 observations had total concentrations above the established upper limit with particles > 0.100 μm exhibiting concentrations above 10^5 $cm^{-3}$and the remaining 39 observations showed zero concentrations for ultrafine particles, thus implying that these measurements were affected by instrument malfunction.

The next step was to consider the differences between weekday and weekend measurements. In general, there were somewhat bigger gaps in data for the weekend. In addition, Morawska *et al.* (2002) found that in Brisbane particle number was 47% higher on weekdays than on weekends. Therefore it was considered that weekend data was of less interest in achieving the aim of this work and therefore excluded from the database used for the analysis.

The final step in the data preparation was the analysis of the missing information. Treatment of missing information ranges from deletion of missing cases to data

imputations. This study aimed to model the existing data and therefore a different approach was considered more appropriate. A common procedure in missing value analysis is testing the assumption that the data is missing completely at random (MCAR), which means that there is no link between missing data and some explanatory variable. Particle number was expected to exhibit daily, weekly and annual fluctuations and therefore the data would be MCAR if the missing information was independent of the time parameters associated with these fluctuations. Pre-1998 missing data was clearly dependent on time of the day and therefore the MCAR assumption had to be redefined. A less stringent and therefore more appropriate approach was the assumption of data missing at random (MAR), indicating no link between a data subset and some control variable. The hypothesis that missing data was independent of month of measurement was tested using the procedure in section 2.5.1, taking the number of unavailable measurements per monthly interval as the dependent variable and month of the year as the group variable.

2.5.3 Time series analysis methodology

The most salient features of a time series are the trend, seasonal and irregular components. The trend and seasonal components were extracted by expressing the data as function of time through univariate and multivariate linear regression models respectively. The results from missing value analysis showed that the data were MAR and therefore it was appropriate to divide it into monthly intervals, using the mean concentrations as the observations. The parameter estimates for the models were obtained through weighted least squares. Variances were incorporated as weights so that the models would allocate less weight to larger variance estimates.

For the trend models, a dummy variable representing the number of months since first observation (t) was added to the database and the estimates were regressed on time through the equation (Chatfield, 1989):

$$X_t = \alpha + \beta t + \varepsilon_t \quad \textbf{(4)}$$

where X_t represents the actual data point at time t, α the intercept of the equation, β the slope, and ε_t denotes a random error term with mean around zero.

For the seasonal model, a series of dummy Boolean variables representing the months of January (i = 1) to November (i = 11) were inserted as regressors, following the procedure shown in Makridakis *et al*. (1983), so that the models took the form:

$$X_t = e_t + b_0 + \sum_{i=1}^{11} b_i D_i \quad \textbf{(5)}$$

Where e_t is the estimated residual, b_0 the intercept, D_i the Boolean regressors and b_i their coefficients. The December effect is measured by the intercept while the effect of other months is measured by the difference between the intercept and the coefficients of the regressors.

The residuals (differences between observed and fitted values) were extracted and analysed in order to investigate the presence of regularities yet to be modelled. Residual analysis included time plots, correlogram analyses and normal probability plots.

3. Results

3.1. Short-term variations in particle number

Normality of the distribution of the hourly means and homogeneity of the hourly variances were checked through Levene's tests. The results indicated that there were significant deviations from the above conditions at the 99% confidence level (p <

0.01) and therefore it was necessary to transform the original data through their natural logarithms. The means and standard variation of the transformed values were then calculated and plotted against time of the day, as shown in Figures 2(a) and 2(b) respectively.

Figure 2(a) shows that, for most of the day, the N_{15-30} remains the largest contributor to the total count, followed by the N_{30-50} and in turn larger size classes. An exception occurs between midnight and 4:00, where the largest contribution comes from the N_{50-100}, followed by the N_{30-50}. The sum of the mean concentrations is calculated by adding the inverse function of the natural logarithms of the concentrations of each class (i.e. $N_{15-630} = \sum e^{\ln(N_{p-q})}$, where N_{p-q} is the number concentration of each size class p-q). Using this formula, total particle number reaches maximum number concentration of 17.3 x 10^3 cm^{-3} at 7:00 hrs and a minimum of 5.14 x 10^3 cm^{-3} at 16:00 hrs.

Figure 2(a) also shows that although particle number declines sharply after the morning peak, the decline is less pronounced for the fraction > 0.100 μm ($N_{100-630}$), especially at around midday. Particle levels then increase gradually between 18:00 and 21:00, remaining more or less stable at about 5.5 x $10^3 cm^{-3}$ until the next morning peak.

Figure 2(b) shows that daily variability in hourly standard deviations is very stable and the size classes follow more or less parallel patterns. The only exception is the $N_{300-630}$, which shows a higher and broader range of variance and several peaks during the day, reaching its highest peak at around 9:00, coinciding with a drop in variance for the smaller size classes. The reason for the daily pattern of variance in the $N_{300-650}$

being different is likely to be linked to the fact that its concentration range is on to two orders or magnitude lower than the concentrations of the other size classes.

In general, as it can be seen in Figure 2(a), all size classes follow more or less the same daily pattern of variation. In the first instance, a one-way ANOVA test was applied to the hourly mean of total particle number concentration to test the hypothesis that particle number varied with respect to time of the day. The results indicated variations in particle number associated with time of the day ($p < 0.01$ for all size classes). Post hoc pairwise comparisons indicated the presence of four data subsets: morning rush (6:00-10:00), midday (11:00-15:00), afternoon rush (16:00-19:00) and night-time (20:00-5:00). The application of ANOVA and pairwise comparisons to the other size fractions produced similar results, confirming that all size classes exhibited the same daily variability, pointing out to the same source as the origin of these particles, which in this case was predominantly traffic.

The interpretation of the results included analyses of the daily variations in traffic flow and wind direction, as was shown in Figures 3(a) and 3(b) respectively. Wind originating from the freeway prevails between midnight and the morning rush, which occurs around 6:00-9:00, causing the observed peak during these hours. In the afternoon rush (around 16:00-17:00), the wind direction is reversed, moving from the station towards the freeway, thereby substantially reducing the amount of the fresh traffic emissions and bringing to the station well-mixed air from the CBD.

3.2. Time series results

The most complete data subsets corresponded to the morning and afternoon measurements. These two data subsets represented two different types of emissions, as discussed above, affected by fresh emissions from the freeway in the morning and

well-mixed urban air in the afternoon. Therefore the understanding of their nature is necessary for the interpretation of the trend model outputs. The summaries of these two subsets are shown in Table 1. The statistics include the mean, standard deviation, minimum and maximum values, and their quartiles. Figures include the NMD for the total size range (in nm), as well as the number concentrations (cm^{-3}) of each of the chosen size classes and the total.

The next step was to prepare the morning and afternoon data subsets for time series analyses. Unlike the hourly estimates of the log-transformed data, the corresponding log-transformed monthly estimates were not normally distributed. The best approximation to normal distribution was obtained by logarithmic transformation of the monthly mean and variance estimates obtained from the original measurements.

Figure 4 presents the time series and corresponding trend components of (a) total particle number concentration, (b) $N_{15\text{-}30}$, (c) $N_{30\text{-}50}$, (d) $N_{50\text{-}100}$, (e) $N_{100\text{-}300}$, (f) $N_{300\text{-}630}$ and (g) NMD. Student t-tests were applied to the trends and the outputs indicated that only the morning trends were significant at the 95% confidence level. The morning trend accounted for 11.8%, 1.6%, 8.9%, 19.6%, 18.5%, 7.6% and 17.0% of the total variance for each series in (a) to (g) respectively.

The results showed that, in the morning, total particle number ($N_{15\text{-}630}$) increased from 7.5 to 15.4 x 10^3 cm^{-3} (i.e. by 105.3%). Of these, the ultrafine fraction ($N_{15\text{-}30}$-$N_{50\text{-}100}$) increased from 6.5 to 11.8 x 10^3 cm^{-3}, which represents an 81.5% increase.

F-tests were applied to the seasonal models and the results indicated that they were statistically insignificant ($p > 0.05$). The only exception was for the morning NMD, which showed a weak seasonal component accounting for 16.7% of its variance. In a further attempt to identify a seasonal pattern, autocorrelation plots were applied to

each series. Figure 5 shows the autocorrelation plot of (a) total particle number and (b) NMD. The remaining autocorrelations were very similar to that in (a), particularly in terms of periodic behaviour, and are therefore not shown in this paper.

4. Discussion and conclusions

Analysis of the submicrometre particle number data collected over a five-year period clearly shows the same daily variation pattern on weekdays, for all size classes used in this study (Figure 2). The statistical tests indicated a time of the day effect upon particle number, with the maximum concentrations occurring around 6:00-7:00 hrs and minimum concentrations at 16:00-17:00. The range of the average total particle number concentrations found in this study was 5.14-17.3 x 10^3 cm^{-3}. This range compares well with the concentrations reported for less polluted European cities and Santiago de Chile, as presented in (Figure 6), which shows the average daily particle number concentrations in selected cities around the world. The average concentrations measured in Brisbane's are much lower than those reported for cities like Birmingham, Rome or the supersites in Detroit and Fresno.

Comparisons with the daily variation pattern in other cities (eg. Harrison *et al.*, 1999; Junker *et al.*, 2000) show that there are differences between Brisbane and those cities. In Basel, Switzerland (Junker *et al.*, 2000), peak concentrations were obtained during the morning and evening and rush hours whereas in Birmingham, UK, particle number peaked around midday (Harrison *et al.*, 1999). Table 1 shows that at the Brisbane monitoring site particle number concentration levels in the morning were, on average, 1.5 times higher than in the afternoon. The interpretation of this daily pattern in concentrations is that it is affected by the daily patterns of traffic and wind direction, as shown in Figure 3. The peak observed in the morning rush hour is affected by emissions coming directly from the freeway to the station whereas the

lack of a corresponding peak in the afternoon rush is explained by the reversal of wind direction moving towards the freeway. This interpretation is based on the results obtained by Hitchins *et al*. (2000), who found that at a distance of 150 m particle number concentration was significantly affected, but decreased by 50% of its value when the wind was blowing directly from the road (in this study the distance was 100 m) whereas particle number was similar to background urban concentrations even at the closest point to the road when the wind blew in the opposite direction.

While there are differences in particle concentration levels between the morning and the afternoon, there were no significant differences in NMD, suggesting similar number size distributions and hence a predominance of vehicle emissions in the air affected by the fresh emissions from the road as well as in urban-background atmosphere. This assertion is based on the findings of Morawska *et al.* (1999), who showed that urban-influenced aerosols had a similar number size distribution as traffic-influenced aerosols, although the peak was not as clearly defined thus indicating the ageing of well-mixed emissions from many sources and a strong influence of traffic.

In relation to long-term trends in particle concentrations, this study found that only the morning trend was significant and that during this time of the day total particle number increased from 7.5 to 15.4 x 10^3 (or 105.3%). In this study, submicrometre particles were divided into different size classes in order to derive more information on their changes in number size distribution. Our results show that although the $N_{15\text{-}30}$ remained the biggest contributor to the total count, its contribution decreased from 38.7% to 26.7% whereas the contribution of $N_{50\text{-}100}$ and the $N_{100\text{-}300}$ increased from 21.8% to 28.7% and from 11.6% to 16.6% respectively. This resulted in a shift in NMD as shown in Figure 3 (g). Nevertheless, the increase was only relative to the

number concentration of each size class. NMD increased from 42.6 to 46.0 nm, which represents only a 3.4 nm increase (7.9%) and remains within the NMD range for vehicle exhaust-emissions (Morawska *et al.*, 1998b; 1996).

In order to shed more light on the reasons underlying differences in particle number behaviour in the morning and afternoon, trend analysis for other air quality and meteorological parameters, including B_{sp}, PM_{10}, Wind Speed, CO, NO and NO_2 were conducted. The results are summarised in Table 2. The trends were calculated from the monthly mean values and weighted (except for CO) against their corresponding monthly variances using least squared regression models. All values, except those for wind speed were transformed using logarithmic scale to normalize their distributions and stabilize their variances. Only some of the trends were found to be statistically significant. In particular, for the morning trends, only PM_{10} shows a statistically significant increasing trend. However, unlike particle number, its afternoon trend also increases thus confirming the independence between particle mass and particle number (Morawska *et al.*, 1998b). During this period, major construction projects took place in and around campus thereby influencing the observed trends in particle mass. B_{sp}, in contrast, shows generally decreasing trends. A report by the Environmental Protection Agency (Neale and Wainwright, 2001a) indicates that during the drought periods of 1993-1996 and 2000 dry conditions increased the frequency and intensity of wildfires and biomass burning thereby increasing the mass concentrations of fine particles (< 2.5 μm), but during 1997-1999 their concentrations remained low therefore influencing the observed B_{sp} trends.

Of the gaseous data, only CO and afternoon NO show significant trends. Being directly emitted into the air, these emissions show long-term reductions in the morning and afternoon. The CO and NO trends are likely to be the result of

technological improvements in vehicle design and the addition of catalytic converters (Neale and Wainwright, 2001a). The effects of these changes upon particle emissions, however, are not yet fully understood and an in-depth analysis of these effects is beyond the scope of the present investigation. Although the trends in primary gas emissions appear to be uncorrelated with particle number emissions, due to the different nature of these pollutants, both gases and particles are affected by trends in the vehicle fleet. Although there has been an increase in vehicle usage in the Southeast Queensland region (Neale and Wainwright, 2001b), there is no evidence to suggest that traffic flow in the freeway has also increased. The trend in morning particle emissions is likely to be linked to changes in the composition of the freeway traffic. Unfortunately, since there was no vehicle speciation data available during the project, this hypothesis cannot be verified.

With regard to particle seasonality, no seasonal pattern was identified in this study. . As it has been mentioned earlier, at times, mainly during the winter months between July and September, biomass burning could be an important contributor to the particle count. Vegetation burning-influenced aerosols have modal diameter around 0.06 μm, which is 0.02 μm larger than particles associated with traffic and urban background emissions (Morawska *et al.*, 1999). Birmili *et al*. (2001) found that typical aerosol number size distributions are attributable to their air mass origin. Therefore, if all other factors were held constant, NMD would be expected to increase during these months, especially in the afternoon. The results, however, do not support this.

Hussein *et al.* (2004) observed that the seasonal behaviour in particle number in Helsinki was inversely related to that of air temperature. Consistent to this pattern, many overseas urban studies have found higher number concentrations, specially at the lower end of the size range, in winter than in summer, independent of other

periodic variations (Cabada *et al.*, 2004; Ebelt *et al.*, 2001; Wehner and Wiedensohler, 2003). Hussein *et al.* (2006) found that ambient temperature and local wind conditions were the most influential factors controlling particle number, but this influence was greater on particles larger than ultrafine. In contrast, the results obtained by Tunved *et al*. (2003) at five non-urban stations in Sweden and Finland showed higher concentrations in summer and spring than in winter. This suggests that in urban environments the association between particle emissions and meteorological conditions is influenced by social factors including summer holidays and household heating in winter. No study of this type has been conducted in a similar environment to that of Brisbane. While the studies above were conducted in areas where there are significant meteorological differences between summer and winter, resulting in significant variations in human life style patterns, in subtropical Brisbane there are much smaller differences between the seasons.

Acknowledgement

The submicrometre particle size distribution data used in this study was collected by Drs. Stephen Thomas and Graham Johnson. Other air quality and meteorological data were supplied by Drs. David Wainwright and Donald Neale, from the Environmental Protection Agency, to whom we are grateful for their ongoing support and advice during the course of this study. The authors also wish to extend their appreciation to colleagues of the International Laboratory of Air Quality and Health (ILAQH) for their support and advice during different stages of the project.

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Figure Captions

1. Monitoring site (AMRS) in relation to the CBD and Southeast Freeway.
2. Particle number dependence on time of the day as measured at QUT AMRS between July 1995 and November 2000: (a) Mean hourly particle number and (b) hourly standard deviations.
3. (a) Daily variation in vehicle traffic on weekdays on the Southeast Freeway. The datapoints represent mean hourly traffic counts (b) Daily pattern of predominant wind direction around QUT AMRS. The datapoints represent the modal wind direction values.
4. Time series plots of monthly mean particle number concentrations in the morning peak (solid line) and afternoon peak (dashed line), over the period July 1995-November 2000 on weekdays in the following size ranges (μm): (a) 0.015-0.630, (b) 0.015-0.030, (c) 0.030-0.050, (d) 0.050-0.100, (e) 0.100-0.300, (f) 0.300-0.630 and (g) monthly mean NMD. All the data has been transformed using the natural logarithmic scale. Successive observations are joined by straight lines.
5. Auctocorrelation functions of morning and afternoon (a) total particle number and (b) number median diameter.
6. Mean particle number concentration levels obtained in selected cities around the world. The figures indicate average values of all the estimates provided in each study.

Table 1

	Mean	Std. Dev.	Min.	Max.	Percentiles		
					25th	50th	75th
Morning Peak (6:00-10:30)							
NMD (nm)	44.4	13.1	22.2	197.6	35.1	42.6	50.8
*N_{Total} (cm^{-3})	$1.28x10^4$	$1.98x10^4$	13.1	$1.99x10^5$	$4.62x10^3$	$8.34x10^3$	$1.39x10^4$
N_{15-30} (cm^{-3})	$4.07x10^3$	$6.91x10^3$	1.3	$8.13x10^4$	$1.33x10^3$	$2.41x10^3$	$4.46x10^3$
N_{30-50} (cm^{-3})	$3.02x10^3$	$4.48x10^3$	2.9	$7.59x10^4$	$1.09x10^3$	$2.04x10^3$	$3.42x10^3$
N_{50-100} (cm^{-3})	$3.12x10^3$	$5.58x10^3$	5.7	$1.78x10^5$	$1.09x10^3$	$2.04x10^3$	$3.62x10^3$
$N_{100-300}$ (cm^{-3})	$2.11x10^3$	$5.37x10^3$	3.3	$1.84x10^5$	$0.64x10^3$	$1.23x10^3$	$2.12x10^3$
$N_{300-630}$ (cm^{-3})	$0.47x10^3$	$2.52x10^3$	0.0	$3.16x10^4$	42.0	82.6	$0.15x10^3$
Afternoon Peak (16:00-19:30)							
NMD (nm)	44.4	13.5	21.6	249.6	35.8	42.1	50.2
*N_{Total} (cm^{-3})	$8.67x10^3$	$1.85x10^4$	7.6	$1.97x10^5$	$2.76x10^3$	$4.67x10^3$	$8.21x10^3$
N_{15-30} (cm^{-3})	$2.83x10^3$	$6.75x10^3$	2.1	$7.8x10^4$	$0.69x10^3$	$1.36x10^3$	$2.66x10^3$
N_{30-50} (cm^{-3})	$2.08x10^3$	$3.84x10^3$	2.0	$9.20x10^4$	$0.72x10^3$	$1.18x10^3$	$2.22x10^3$
N_{50-100} (cm^{-3})	$1.97x10^3$	$3.94x10^3$	2.1	$7.62x10^4$	$0.66x10^3$	$1.14x10^3$	$1.99x10^3$
$N_{100-300}$ (cm^{-3})	$1.35x10^3$	$3.40x10^3$	1.0	$3.89x10^4$	$0.41x10^3$	$0.68x10^3$	$1.18x10^3$
$N_{300-630}$ (cm^{-3})	$0.44x10^3$	$2.75x10^3$	0.0	$3.18x10^4$	24.4	46.9	91.0
Morning/Afternoon Ratios							
NMD (nm)	1.000	0.970	1.03	0.792	0.980	1.012	1.012
*NTotal (cm-3)	1.474	1.068	1.72	1.010	1.675	1.785	1.696
N15-30 (cm-3)	1.436	1.025	0.62	1.042	1.919	1.769	1.676
N30-50 (cm-3)	1.456	1.169	1.45	0.824	1.506	1.729	1.538
N50-100 (cm-3)	1.581	1.418	2.71	2.331	1.647	1.831	1.815
N100-300 (cm-3)	1.561	1.579	3.30	4.718	1.559	1.820	1.798
N300-630 (cm-3)	1.063	0.915	-	0.994	1.721	1.761	1.698

Summary statistics of particle number concentration and number median diameters measured at QUT monitoring station during the morning and afternoon peak traffic hours on weekdays from July 1995 to November 2000.

Table 2

Parameter	Morning	Afternoon
B_{sp}	$^{*}6.12x\ 10^{-6}m^{-1}$ (31.8%) decrease	$^{*}5.62\ x\ 10^{-6}m^{-1}$ (30.5%) decrease
PM_{10}	$^{*}0.45\ \mu g\ m^{-3}$ (13.4%) increase	$^{**}0.18\ \mu g\ m^{-3}$ (5.38%) increase
Wind Speed	$0.14\ m.s^{-2}$ (26.6%) decrease	$^{**}0.47\ m.s^{-1}$ (32.1%) decrease
CO	*0.45 ppm (62.5%) decrease	*0.47 ppm (69.0%) decrease
NO	$8.79\ x\ 10^{-2}$ pphm (2.38%) decrease	**5.57 pphm (30.5%) decrease
NO_2	No change	0.11 pphm (7.84%) increase

*Trend is significant at the 99% confidence level

**Trend is significant at the 95% confidence level

Summary of the morning and afternoon trends of B_{sp}, PM_{10}, wind speed, CO and NO_x data measured at QUT AMRS during June 1995-November 2000.

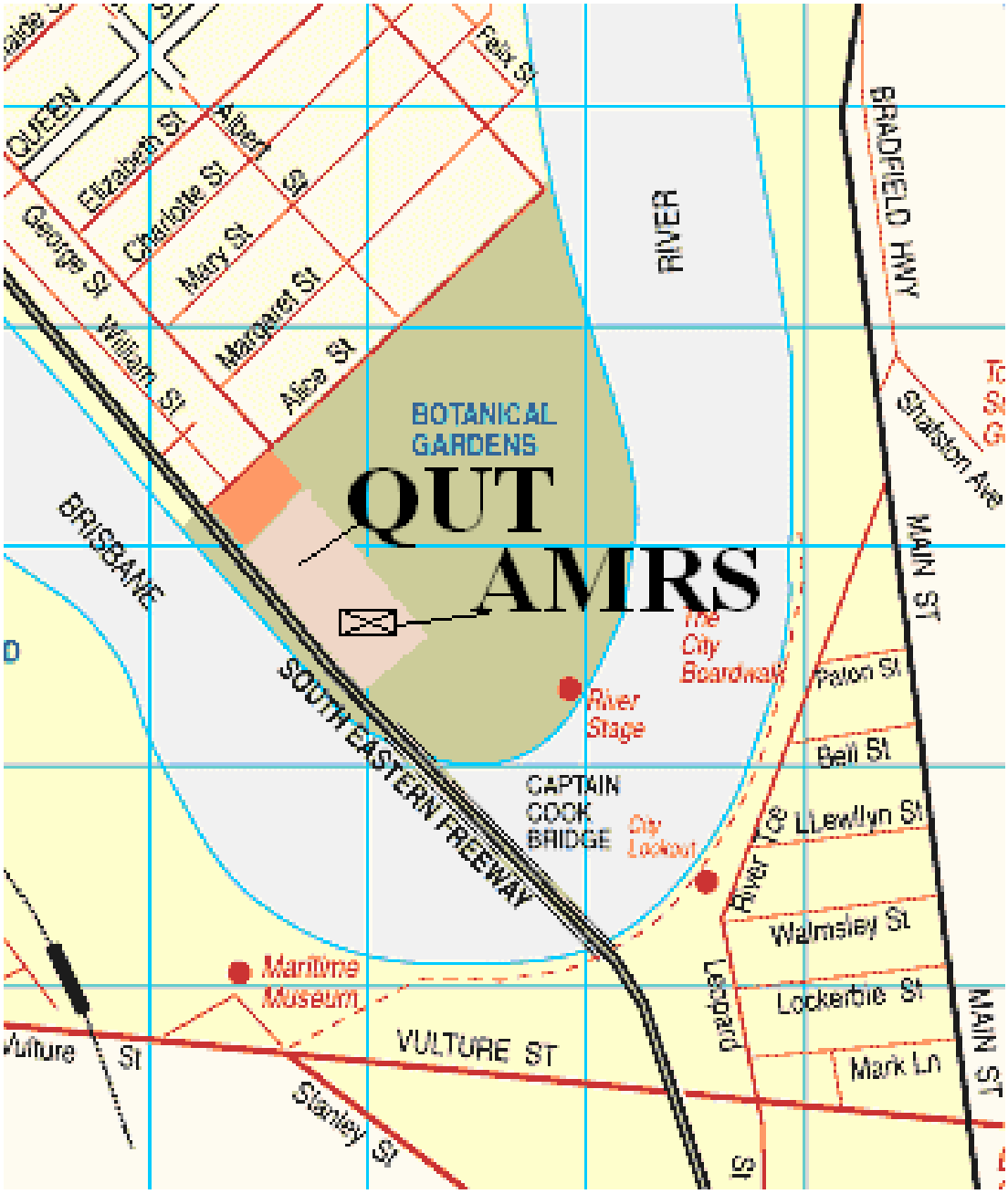
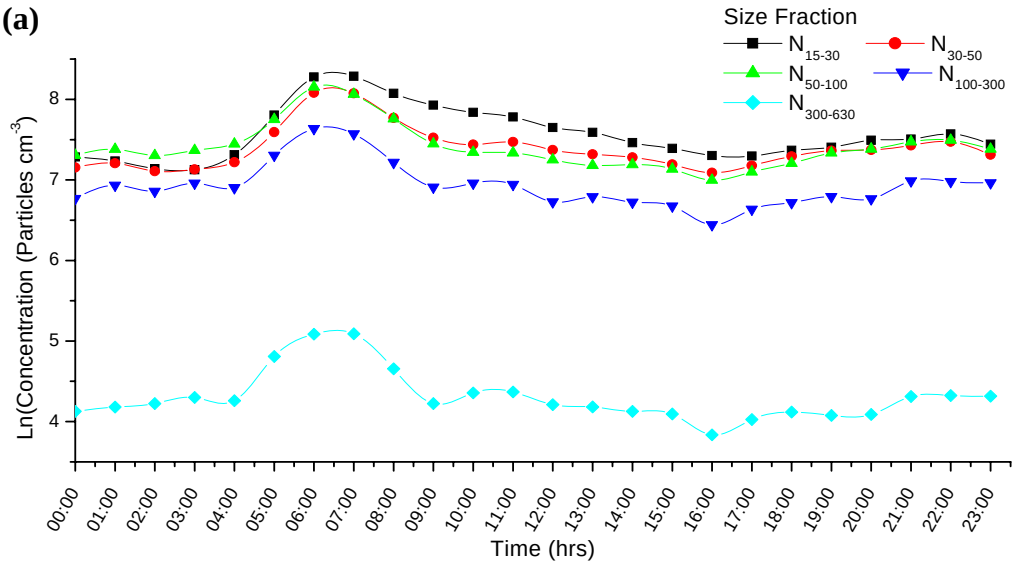

QUEEN
Elizabeth St
Charlotte St
Mary St
Margaret St
Alice St
George St
William St
Albert St
Felix St
RIVER
BRADFIELD HWY
Gladstone Ave
MAIN ST
BOTANICAL GARDENS
QUT
AMRS
BRISBANE
SOUTH EASTERN FREEWAY
The City Boardwalk
River Stage
Paton St
Bell St
CAPTAIN COOK BRIDGE
City Lookout
& Llewellyn St
Walmsley St
Lockerbie St
Mark Ln
Maritime Museum
VULTURE ST
Vulture St
Stanley St
Leopard St

Figure 1

(a)


Size Fraction
N15-30
N30-50
N50-100
N100-300
N300-630
Ln(Concentration (Particles cm-3)
Time (hrs)

(b)

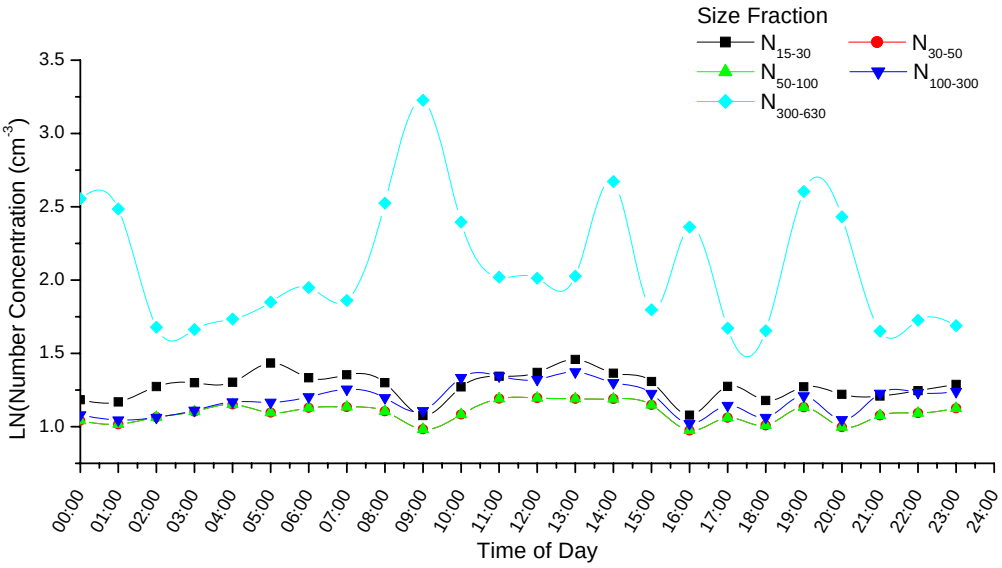
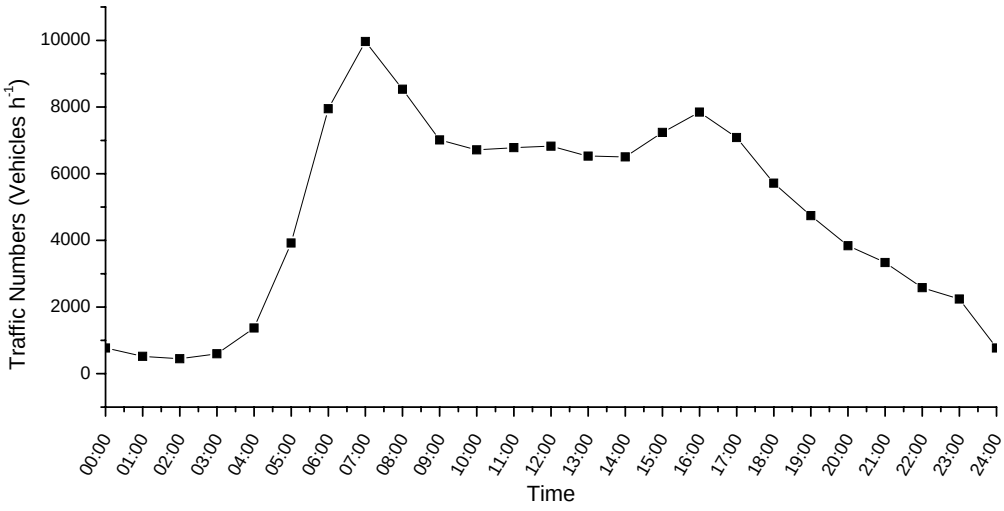

Size Fraction
N15-30
N30-50
N50-100
N100-300
N300-630
LN(Number Concentration (cm-3))
Time of Day

Figure 2

(a)


Traffic Numbers (Vehicles h-1)
10000
8000
6000
4000
2000
0
00:00
01:00
02:00
03:00
04:00
05:00
06:00
07:00
08:00
09:00
10:00
11:00
12:00
13:00
14:00
15:00
16:00
17:00
18:00
19:00
20:00
21:00
22:00
23:00
24:00
Time

(b)

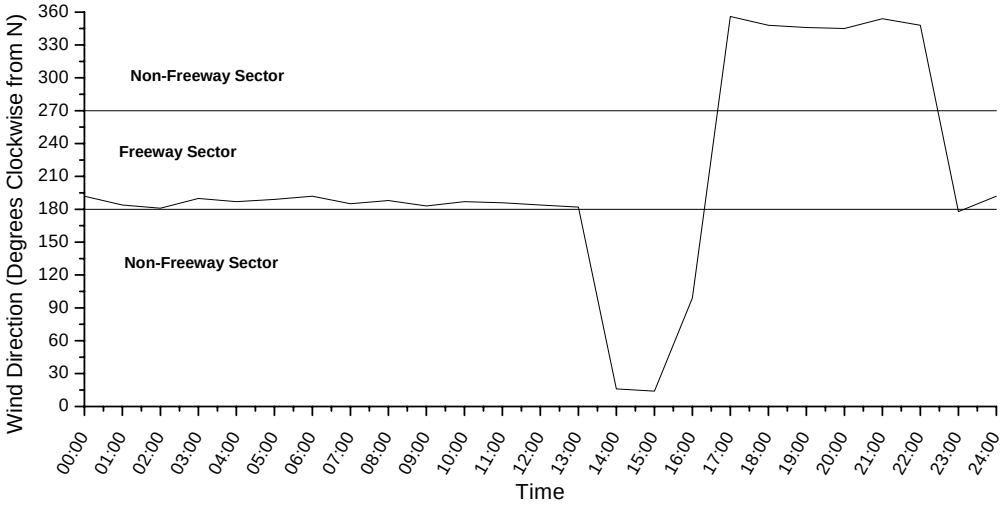
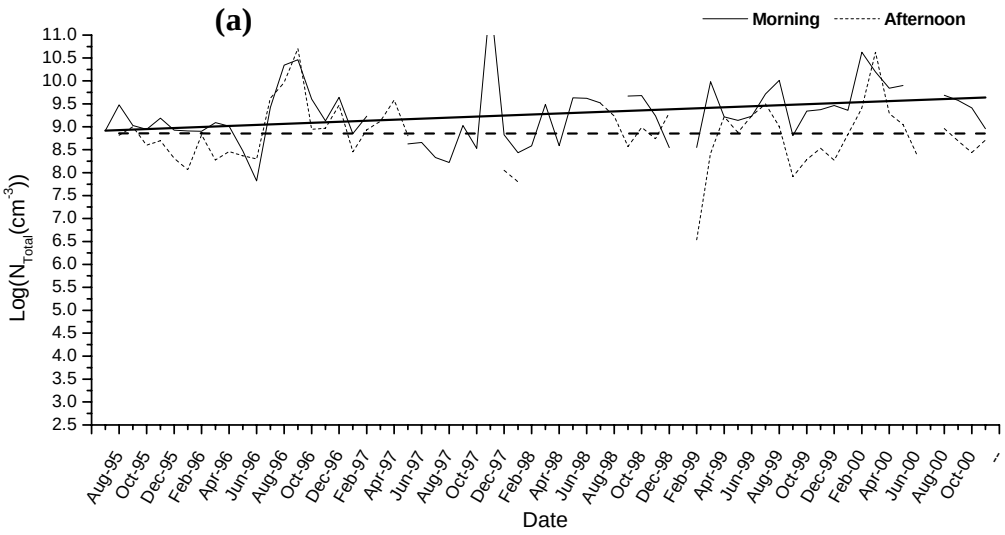
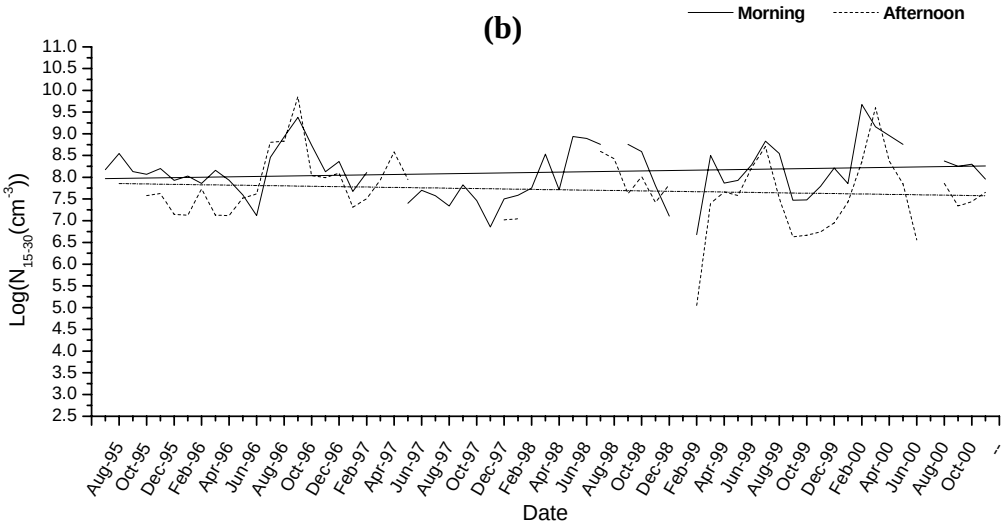
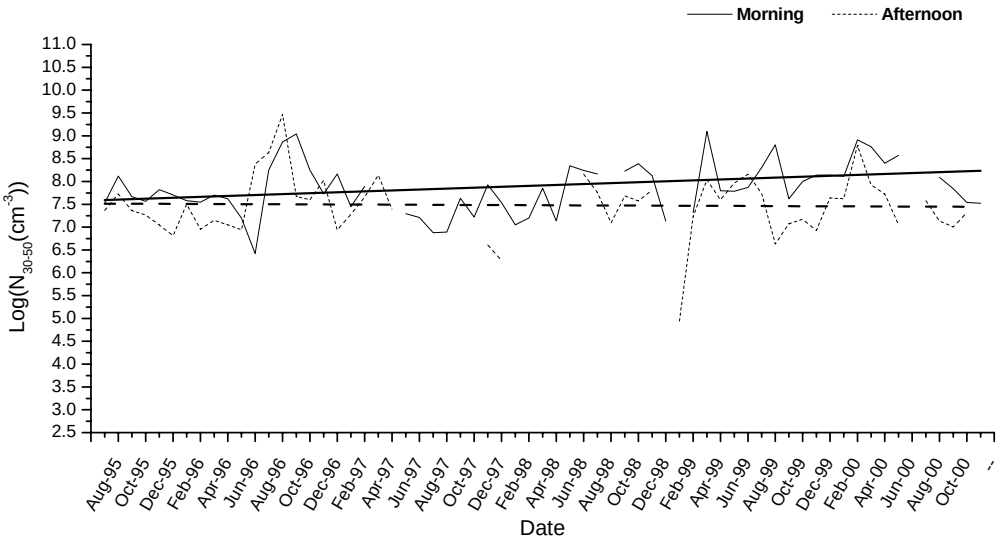

Wind Direction (Degrees Clockwise from N)
360
330
300
270
240
210
180
150
120
90
60
30
0
Non-Freeway Sector
Freeway Sector
Non-Freeway Sector
00:00
01:00
02:00
03:00
04:00
05:00
06:00
07:00
08:00
09:00
10:00
11:00
12:00
13:00
14:00
15:00
16:00
17:00
18:00
19:00
20:00
21:00
22:00
23:00
24:00
Time

Figure 3

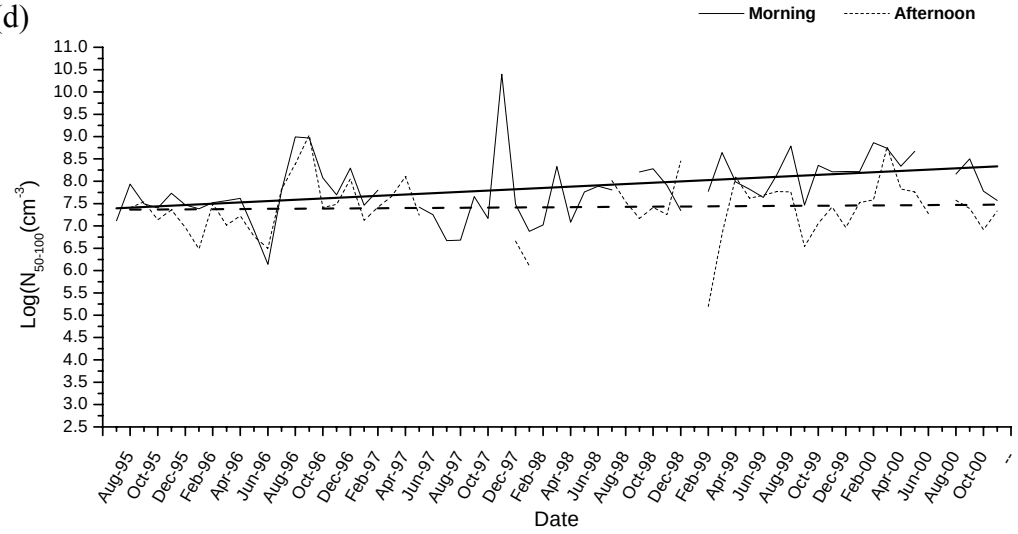

(a)
Morning
Afternoon
Log(N_Total(cm^-3))
Date


(b)
Morning
Afternoon
Log(N_15-30(cm^-3))
Date

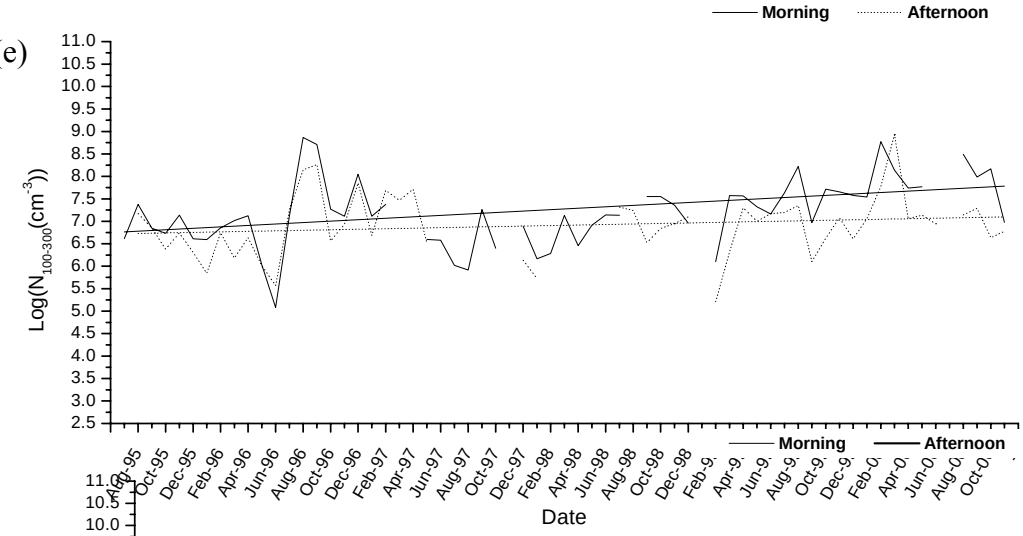
(c)


Morning
Afternoon
Log(N_30-50(cm^-3))
Date

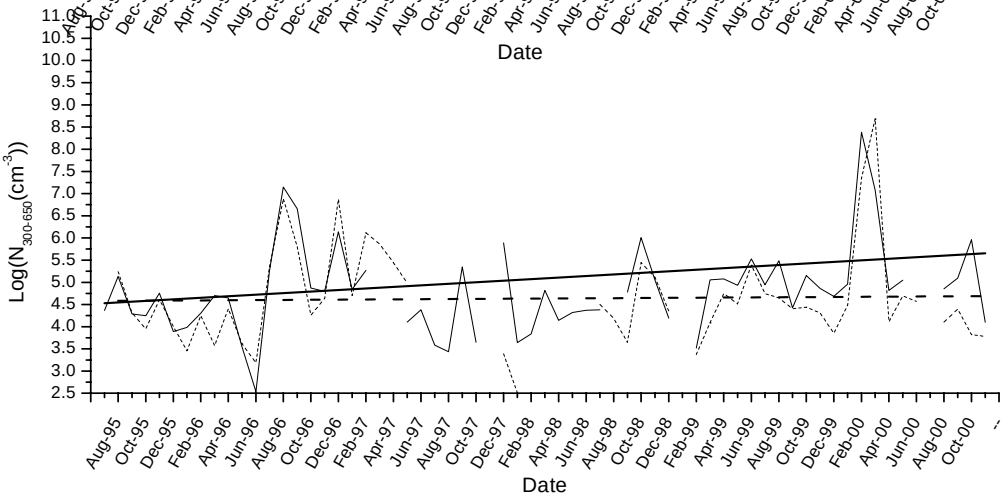
(d)


Morning
Afternoon
Log(N50-100(cm-3)
Date

(e)


Morning
Afternoon
Log(N100-300(cm-3))
Date

(f)


Log(N300-650(cm-3))
Date

(g)

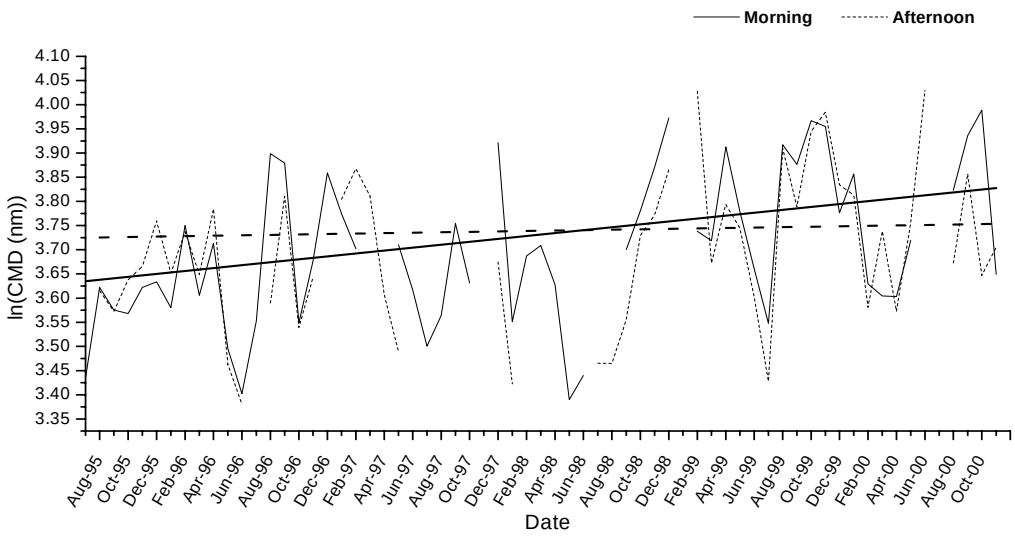
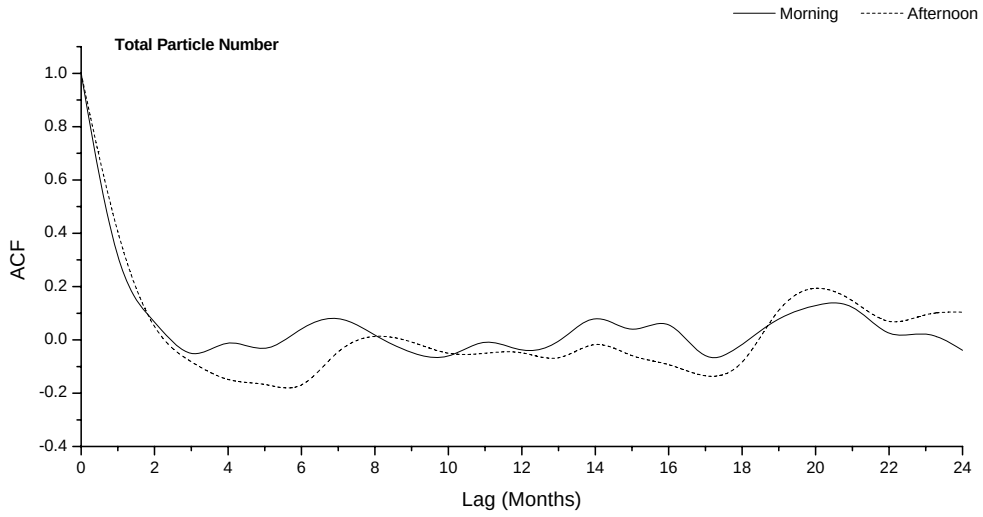

Morning
Afternoon
ln(CMD (nm))
4.10
4.05
4.00
3.95
3.90
3.85
3.80
3.75
3.70
3.65
3.60
3.55
3.50
3.45
3.40
3.35
Aug-95
Oct-95
Dec-95
Feb-96
Apr-96
Jun-96
Aug-96
Oct-96
Dec-96
Feb-97
Apr-97
Jun-97
Aug-97
Oct-97
Dec-97
Feb-98
Apr-98
Jun-98
Aug-98
Oct-98
Dec-98
Feb-99
Apr-99
Jun-99
Aug-99
Oct-99
Dec-99
Feb-00
Apr-00
Jun-00
Aug-00
Oct-00
Date

Figure 4


Morning
Afternoon
Total Particle Number
ACF
1.0
0.8
0.6
0.4
0.2
0.0
-0.2
-0.4
0
2
4
6
8
10
12
14
16
18
20
22
24
Lag (Months)

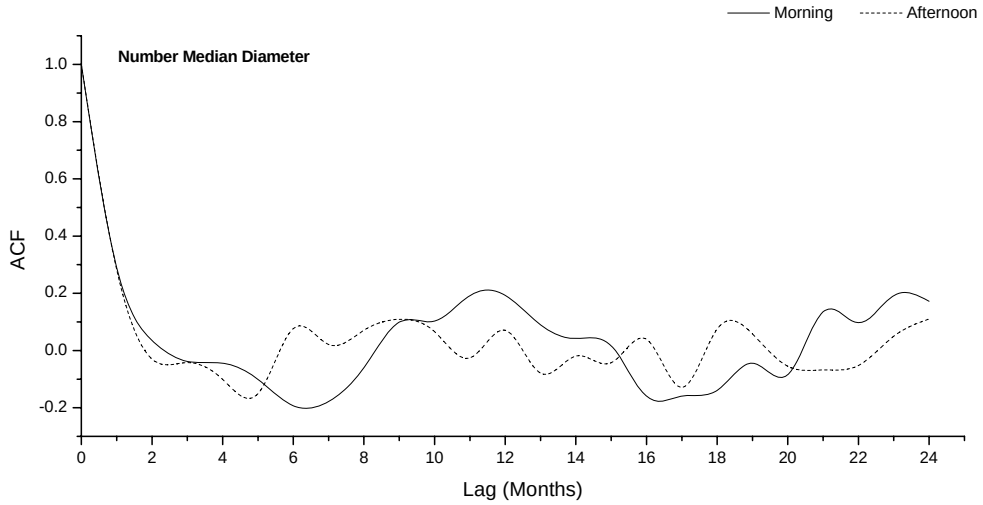

Morning
Afternoon
Number Median Diameter
ACF
1.0
0.8
0.6
0.4
0.2
0.0
-0.2
0
2
4
6
8
10
12
14
16
18
20
22
24
Lag (Months)

Figure 5

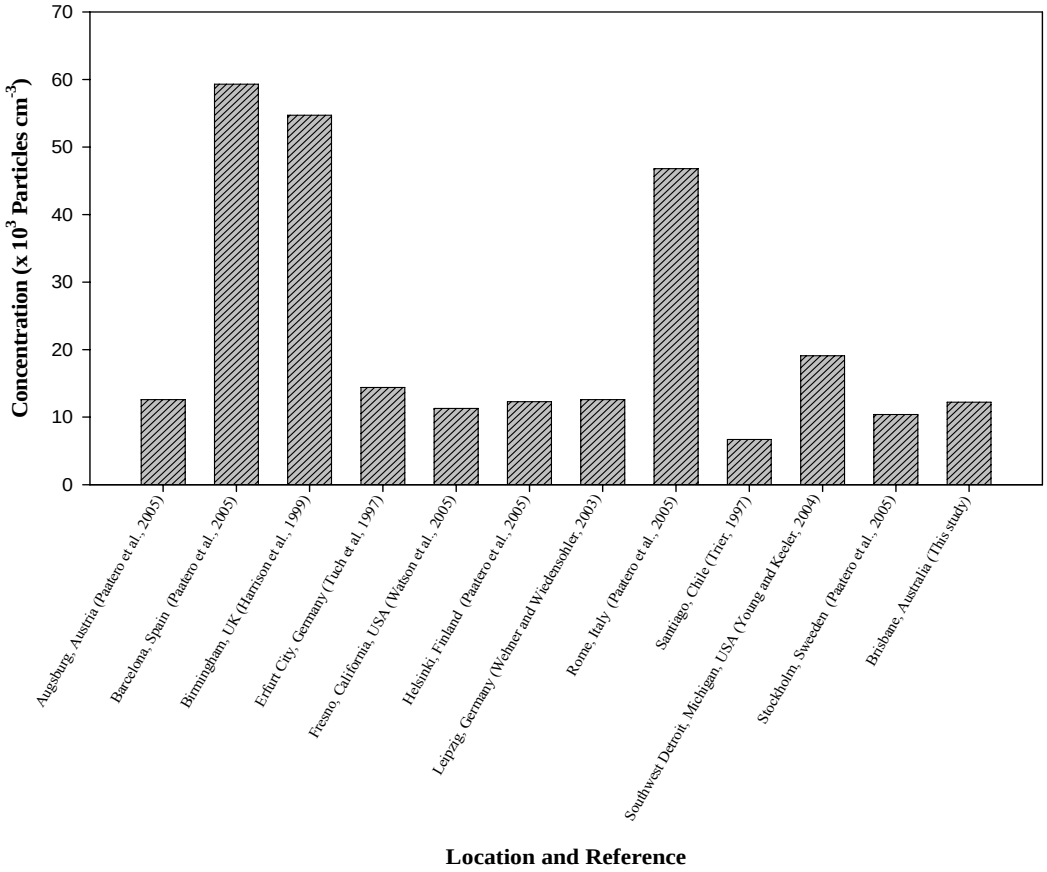

Concentration (x 10^3 Particles cm^-3)
70
60
50
40
30
20
10
0
Augsburg, Austria (Pastero et al., 2005)
Barcelona, Spain (Pastero et al., 2005)
Birmingham, UK (Harrison et al., 1999)
Erfurt City, Germany (Tuch et al, 1997)
Fresno, California, USA (Watson et al., 2005)
Helsinki, Finland (Pastero et al., 2005)
Leipzig, Germany (Wehner and Wiedensohler, 2003)
Rome, Italy (Pastero et al., 2005)
Santiago, Chile (Trier, 1997)
Southwest Detroit, Michigan, USA (Young and Keeler, 2004)
Stockholm, Sweeden (Pastero et al., 2005)
Brisbane, Australia (This study)
Location and Reference

Figure 6