



Spatial variation in particle number size distributions in a large metropolitan area

J. F. Mejía, L. Morawska, K. Mengersen

► To cite this version:

J. F. Mejía, L. Morawska, K. Mengersen. Spatial variation in particle number size distributions in a large metropolitan area. *Atmospheric Chemistry and Physics Discussions*, 2007, 7 (6), pp.17147-17177. hal-00303189

HAL Id: hal-00303189

<https://hal.science/hal-00303189>

Submitted on 18 Jun 2008

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

**Spatial variation in
particle number size
distributions**

J. F. Mejía et al.

Spatial variation in particle number size distributions in a large metropolitan area

J. F. Mejía¹, L. Morawska¹, and K. Mengersen²

¹International Laboratory of Air Quality and Health, Queensland University of Technology,
GPO Box 2434, Brisbane, Qld 4001, Australia

²School of Mathematical Sciences, Queensland University of Technology, Australia

Received: 30 August 2007 – Accepted: 24 October 2007 – Published: 26 November 2007

Correspondence to: L. Morawska (l.morawska@qut.edu.au)

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Abstract

Air quality studies have indicated that particle number size distribution (NSD) is unevenly spread in urban air. To date, these studies have focussed on differences in concentration levels between sampling locations rather than differences in the underlying geometries of the distributions. As a result, the existing information on the spatial variation of the NSD in urban areas remains incomplete. To investigate this variation in a large metropolitan area in the southern hemisphere, NSD data collected at nine different locations during different campaigns of varying duration were compared using statistical methods. The spectra were analysed in terms of their modal structures (the graphical representation of the number size distribution function), cumulative distribution and number median diameter (NMD). The study found that with the exception of one site all distributions were bimodal or suggestive of bimodality. In general, peak concentrations were below 30 nm and NMDs below 50 nm, except at a site dominated by diesel trucks, where it shifted to around 50 and 60 nm respectively. Ultrafine particles (UFPs) contributed to 82–90% of the particle number, nanoparticles (<50 nm) to around 60–70%, except at the diesel traffic site, where their contribution dropped to 50%. Statistical analyses found that the modal structures heterogeneously distributed throughout Brisbane whereas it was not always the case for the NMD. The discussion led to the following site classification: (1) urban sites dominated by petrol traffic, (2) urban sites affected by the proximity to the road and (3) an isolated site dominated by diesel traffic. Comparisons of weekday and weekend data indicated that, the distributions were not statistically different. The only exception occurred at one site, where there is a significant drop in the number of diesel buses on the weekend. The differences in sampling period between sites did not affect the results. The statistics instead suggested variations in traffic composition. However, the relative contribution of petrol vehicle emissions at each site could not be assessed due to the limited traffic information available.

Spatial variation in particle number size distributions

J. F. Mejía et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

1 Introduction

Evidence from medical studies indicates that air pollution is a major contributor to cardiovascular and respiratory health problems (e.g. Hong et al., 2002). Until recently, these studies focussed on the particle mass, which is dominated by the coarse (2.5–10.0 μm) and fine (<2.5 μm) size fractions. Whilst the toxicological importance of these particles cannot be dismissed, it has been suggested that the observed effects could be better explained by the number, rather than the mass, of the ultrafine fraction (<100 nm) (Seaton et al., 1995). In urban air, ultrafine particles (UFPs) dominate the particle number size distribution whereas their contribution to the particle mass is negligible (Morawska et al., 1998a, b; Jaenicke, 1993). Therefore, particle number has become the most widely employed parameter for measuring UFP concentration.

Although they are yet to be incorporated into long-term air quality monitoring systems, since the 1990s submicrometre (<1.0 μm) and UFP number concentrations have been measured in several cities around the world. Typical urban concentrations range in the order of 10^3 – 10^5 cm^{-3} (e.g. Buzorius et al., 1999) with the UFPs accounting for >80% of the total particle number (Morawska et al., 1998a, b; Shi et al., 2001). There is wide agreement that vehicles are the principal source of UFPs in urban areas. This has been shown by the strong associations of particle number with vehicle exhaust gases (Morawska et al., 1998b; Wåhlin et al., 2001), variations in particle number consistent with daily patterns of traffic (Harrison et al., 1999) and wind direction (see also Morawska et al., 2002), and higher number concentrations when the wind blows from the road (Hitchins et al., 2000), in areas of higher traffic density (Hussein et al., 2005; Harrison et al., 1999) and during the week, when traffic levels are higher (Morawska et al., 2002).

Particle size is determined by the particle generation process and in turn it influences particle behaviour and other important chemical and physical properties. Therefore, the analysis of particle size distribution can be useful in the interpretation of the properties of the collected samples. Different descriptors of the particle number size distribution

ACPD

7, 17147–17177, 2007

Spatial variation in particle number size distributions

J. F. Mejía et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

EGU

(NSD) have been used to characterize the source signatures of different types of emissions, for instance petrol (Morawska et al., 1998b) and diesel exhaust (Morawska et al., 1998a; CONCAWE, 2001; Virtanen et al., 2004) and the NSD spectra from different environments (e.g. Morawska et al., 1999b; Flocas et al., 2006).

Urban ambient air quality investigations have found variations in the NSD spectra obtained from different locations in the same city (e.g. Hussein et al., 2004, 2005; Salma et al., 2002) but these studies have focussed on the differences in concentration levels rather than the underlying geometries of the distributions. Therefore the existing information on the spatial variation of the NSD in urban areas remains incomplete.

The present investigation aims to analyse the spatial variation of the NSD spectra in a large city in the southern hemisphere, using data collected at nine different locations, and test the hypothesis that the spectra is evenly distributed throughout the area. The objective was to compare the average spectra using different descriptive size distributions parameters, and to evaluate the relative contribution of UFPs and nanoparticles (<50 nm) to the local airborne particle number.

2 Methodology of the investigation

2.1 The city of Brisbane

Brisbane is the capital of the state of Queensland, Australia, and is located in the southeast coast of the state. It has a population of about 1.8 million people, making it the third largest city in the country and the first in terms of population growth. The city straddles the Brisbane River, and is located in a low-lying floodplain, although the area is dotted by large hills reaching up to 300 m.

Brisbane's climate is the sub-tropical humid type (Bureau of Meteorology, 1983). The warm season takes place between November and April, and is characterized by high relative humidity, strong winds with average of 11–21 km h⁻¹ and frequent rain. The cool half of the year (May–October) is dry (0–50% relative humidity) with very low wind speeds thus increasing the persistence time of air pollutants.

Spatial variation in particle number size distributions

J. F. Mejía et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

The rapid population growth and consequently increasing number of vehicles on the road pose a serious challenge to the air quality of the city. In a recent five-year study in the central business district (CBD) of Brisbane (Mejia et al., 2007), we found that although particle number concentration levels compared favourably with other cities around the world, emissions from a nearby freeway resulted in increased total and ultrafine particle concentrations by 105.3% and 81.5% respectively whereas background emissions remained unchanged. These conclusions were based on the available data and their associations with the daily local pattern of wind direction.

Hazard-reduction burning, practiced during the cold season in the outer agricultural and forest areas, is another important contributor to the particle number in the region. On these occasions, the city may be covered in smoke carried through the drainage flows. Commercial and industrial sources include a major airport and a seaport, oil refineries, a power station, and a number of manufacturing industries in the outer suburbs.

2.2 Description of the instrumentation

A TSI model 3394 SMPS was used to measured NSD data in the range 15–700 nm. Details of the operation and calibration of the instrument have been provided in previous publications (e.g. Hitchins et al., 2000; Morawska et al., 1998b) and briefly summarised here. The SMPS consists of a TSI 3071A Electrostatic Classifier (EC), which classifies the particles by their ability to transverse an electric field, and a TSI 3010 Condensation Particle Counter (CPC), which measures their concentration. The SMPS was calibrated before and after the measurement period for the PSD using standard 100 nm latex spheres. The entire system is automated and is equipped with customized software. This software collects and stores sample data, which can then be exported to other applications.

Spatial variation in particle number size distributions

J. F. Mejía et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

2.3 Location of the monitoring sites and sampling procedure

Particle number size distribution and temperature data were collected at nine different locations in Brisbane. Table 1 summarizes the details of these locations and their corresponding databases including the area classification, distance to the CBD, number of recorded cases and the percentage of the retained observations. More details are provided below. Unless otherwise indicated, the measurements were conducted at ground level, at an approximate height of 1.5 and 5–15 m to the road, with the principal road consisting of two lanes travelling in each direction:

1. Queensland University of Technology Air Monitoring Research Station (QUT AMRS) – A joint venture between QUT and the Environmental Protection Agency. The station is located on the sixth floor of a building in QUT Gardens Point campus, at the southern end of the Brisbane CBD. The height of the station is about the same as that of the Southeast Freeway, located approximately 100 m south to the station. The freeway consists of three lanes travelling on each direction. For the purpose of brevity, the site be referred here as QUT.
2. Brisbane Square (BSQ) – Located in the heart of the Brisbane CBD, surrounded by four busy roads 15–20 m wides and buildings 15–30m tall with one building reaching a height of 112 m.
3. Princess Alexandra Hospital (PAH) – Monitoring was conducted on a pedestrian bypass, connecting two five-story buildings, 3.5 m directly above a busy road and a traffic light.
4. Tora St – On Tora St Bridge, 3.5 m directly above the freeway, about 10 km south to the CBD. The freeway consists of three lanes travelling in each direction.
5. Archerfield – In the south-eastern corner of a fast food restaurant car park, and a busy road. A traffic light intersection is located 5 m east to the site

Spatial variation in particle number size distributions

J. F. Mejía et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

6. ANZ sports complex (ANZ) – in the south-eastern corner of the complex. The terrain around the site consists of uncovered loose soil. The sampling location was surrounded two busy roads, <5 m away from the site, intersecting at a traffic light, and a large sports complex.
- 5 7. Garden City (GC) – near the northeast corner of a major shopping centre car park. The shopping centre was bounded by two busy roads to the east and north, intersecting at a traffic light.
8. Mt Gravatt-Capalaba Rd (MGC Rd) – At the intersection of two busy roads in a residential area.
- 10 9. Brisbane Port (BP) – at the end of a bridge connecting the mainland to Fisher-man’s Island, between the road and a railway line. The road consists of one lane travelling on each direction. The area is surrounded by the mouth of the Brisbane River to the West and Moreton Bay to the N and E, both approximately 60 m from the site.
- 15 Monitoring was conducted continuously throughout the day, except at QUT. At this site, prior to 1998, the SMPS was used for regular “grab sampling” of NSD in tripli-cates everyday in the morning (09:30–10:00) and the afternoon and at regular inter-vals throughout the day during periods characterized by notable atmospheric condi-tions such as haziness or hazard reduction burning; from January 1998, the data was
20 collected at hourly intervals on a continuous basis.

The interruptions to monitoring were generally few and short, lasting only a few hours, except at Tora St, which lasted several days, and QUT, which lasted a maxi-mum of three months. These two sites covered the earliest and longest campaigns and therefore the interruptions were mainly caused by equipment maintenance and
25 competing usage priorities.

Spatial variation in particle number size distributions

J. F. Mejía et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

2.4 Preparation of the database for statistical analyses

The lognormal distribution is the most common method of representing the size distribution of atmospheric particles (e.g. Seinfeld and Pandis, 1998) for presenting and analysing the data, and was therefore the selected method for this study. There were occasional variations in the measured size range, so in order to maintain consistency concentrations outside the size range 15–300 nm were removed and the total concentration (i.e. N_{15-300}) was then calculated. The selected size range was based on the results of an earlier Brisbane investigation which found that throughout the day the number concentration of particles >300 nm was between one and two orders of magnitude lower than that of particles <300 nm (Mejia et al., 2007) and therefore their contribution could be considered negligible. The second step was to obtain the relative contribution of each channel to the particle number by dividing its concentration over the N_{15-300} . This was done with the objective of allowing comparisons of the underlying geometry of different NSD spectra, regardless of their actual concentration levels.

The data were divided into major subsets based on wind originating from the principal road (within $\pm 45^\circ$ of the direction perpendicular to the road), wind towards this road, and in some cases, wind from other major physical structures. These subsets were further divided into weekdays and weekends. The aim was to determine whether wind sector or day of the week had an effect on the spectra.

The datasets were subjected to outlier analysis in order to identify and remove extreme values that may lead to an incorrect interpretation of the data. There are several techniques available for outlier identification (e.g. Abu-el-zeet et al., 2002) and their selection depends on the characteristics of the dataset. In the present investigation, outliers were detected using the methodology described in Yu et al. (2004) and briefly summarized here. This is a two-stage graphical and statistical procedure used in the identification of discordant size distributions. The first stage uses the total concentration vs. the corresponding coefficient of variation, both in the logarithmic scale in order to detect size distributions isolated from the main cluster of observations. In the second

Spatial variation in particle number size distributions

J. F. Mejía et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

stage, individual outliers are identified by the Generalized Extreme Studentized Deviate (ESD) Many-Outlier Procedure and the modified z-score (see also Barnett and Lewis, 1994, for more details).

After the outliers were identified and eliminated, the next move was to select those NSD properties to be analysed as well as appropriate data analysis procedure. The modal structure (the graphical representation of the shape of the number size distribution function) has the advantage of providing a detailed description of the structure of the distribution, provided that the diameter size range has been carefully selected in order to maximise the amount of detail of the fraction dominating the particle parameter of interest and that size intervals are of the same width in the same scale. The particle number is overwhelmingly dominated by particles 300 nm (e.g. Eleftheriadis et al., 2006); therefore, the selected size range was therefore considered appropriate for this study.

The Kolmogorov-Smirnov (K-S) test was used to compare the modal distribution obtained from each site. The use and applicability of this test for comparing particle size distributions containing a large number of sizing channels has been described by Heitinbrink et al. (1990) and the details of the application of this technique for measured SMPS data have provided by Morawska et al. (1999a) and further expanded here. The K-S test compares the largest distance between two cumulative distribution functions against a critical value (D_α), which is calculated from the equation:

$$D_\alpha = c(\alpha) \sqrt{\frac{n_1 + n_2}{n_1 n_2}} \quad (1)$$

Where $c(\alpha)$ is the confidence level (α) coefficient, and n_1 and n_2 are the sizes of the two samples. For a 95% confidence level, the value of $c(\alpha)$ is 1.36 (Gibbons and Chakraborti, 2003). The K-S tests were performed using S-Plus 6.1 (2002, Insightful Corp.).

Two other commonly employed parameters are the number median diameter (NMD), which is also calculated from the cumulative distribution function, and the geometric

Spatial variation in particle number size distributions

J. F. Mejía et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

standard deviation (GSD), which measures the width of the distribution. Datasets were compared through one-way ANOVA tests. Homogeneity of variances was analysed using Levene's test. Pairs of means were then compared using least significant difference (LSD) tests in the case of equal variances and Games-Howell tests in the case of unequal variances.

3 Results

3.1 Modal Structure of the NSD

Figure 2 shows the average modal structure of the submicrometre particle NSD obtained from each site. For comparison purposes, the vertical axes have been drawn to the same scale. The figures indicate the percentage of the total concentration in the range 15–300 nm. As shown by these results, at most sites, there were two peaks or modes: one occurring below 30 nm, and a second peak or protrusion suggesting its presence, is observed at above 50 nm. QUT was the exception by having a clearly uni-modal distribution. At BP, the distribution appears to be bimodal although the second peak, at around 50 nm, has a higher concentration than the first peak, which occurs at around 40 nm. At ANZ site, no peak in the lower size range was observed, and the data suggests that it was located below the lower end of the size range measured in this study; also, the second mode is above 100 nm and its sources are yet to be determined.

The NSD data was then used to calculate the cumulative NSD and the resulting spectra are shown in Fig. 3. The objective was to assess the contribution of UFPs and nanoparticles (<50 nm) to the particle number and identify the location of the NMD. The results indicated that UFPs contributed to around 85% of the particle number and nanoparticles to around 60%; at ANZ and Tora St sites the contribution of UFPs and nanoparticles were, respectively, 90% and 70%, probably reflecting their close proximity to the road (<5 m). At BP, UFPs although the contribution of ultrafine particles is

Spatial variation in particle number size distributions

J. F. Mejía et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

consistent with those from the other sites (around 83%), the contribution of nanoparticles drops to 50%, indicating the much higher contribution of diesel trucks. With regard to NMD, the plots show that most NMDs fall in the 40 nm region, except at Tora St and ANZ, where it falls around 30 nm, and BP, where it shifts to nearly 50 nm.

The K-S test compared the distributions between each pair of spectra and the results are shown in Table 3. The tests showed that the spectra differed significantly between all of the sites, thereby indicated that the composition of the emissions is not evenly distributed around the area. Despite these differences, there were three major groups of spectra: the first group corresponded to the sites with peaks at around 30 nm (PAH, Archerfield, Garden City, MGC Rd, QUT and BSQ); the second group was represented by the sites dominated by peaks <25 nm (Tora St and ANZ); and the third group was represented by one site (BP) where particle number peaked at around 50 nm. Although these results hint at differences in the vehicle mix, there was no traffic data available and therefore the effect of traffic composition on size distribution could not be analysed in more detail.

Comparisons between the data subsets for each site indicated that there were no significant differences between wind direction sectors or between weekdays and the weekend ($p>0.05$). Therefore, the extracted spectra above can be considered representative of their corresponding sites. There were minor exceptions, the most significant being at BSQ between weekdays and the weekend ($p=0.02$). The resultant spectra are shown in Fig. 4a and b, respectively. During the week, the distribution is bimodal with peaks at around 30 and 60 nm while on the weekend it is unimodal peaking at around 35 nm.

3.2 Number median diameter (NMD) and geometric standard deviation (GSD)

ANOVA tests applied to NMD found significant differences between sites. Post hoc comparisons indicated that with the exception of PAH, Archerfield, Garden City and MGC Rd, the mean values differed significantly. This was largely expected due to the large sample sizes. However, the magnitudes of most of the differences were found to

Spatial variation in particle number size distributions

J. F. Mejía et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

be very small (maximum of 5 nm) and therefore not practically important, so in practice, they could be grouped as follows: Group 1 was represented by particles in the 40–50 nm size region (the four sites above plus BSQ and QUT); Group 2 comprised those in the 30 nm region (ANZ and Tora St); and Group 3 belonged to the 60 nm region (BP).

5 The mean values for each site are shown in Fig. 5, with the continuous lines used to separate the major groups.

The statistical tests indicated that mean GSD differed very little between sites (mean 1.94–2.02, s.d. 0.12–0.17) except at BP, where it was significantly greater (mean 2.46, s.d. 0.85). In other words, the size distribution showed more or less the same level of polydispersity, except at BP.

3.3 Effect of differences in time period

The NSD measurements were conducted during different campaigns, each having different duration, as indicated in Table 1. The results proved that despite the differences in time period the ranges of obtained values for the location of the peaks and NMD, as well as the contribution of ultrafine particles and nanoparticles to the total concentration, were very narrow. The only exception was BP site, even though the average temperature at this site did not differ from the rest (Table 2). Therefore, it is very clear that the sampling periods did not affect the distributions.

4 Discussion

20 The size distribution of submicrometre particle data, collected at nine different sites in urban atmosphere of Brisbane, in the range 15–300 nm using the lognormal scale, were analysed in terms of their modal structure and NMD, covering a total period of 11 years. Larger particles were excluded from the study as we found in a previous investigation that their concentrations were between one and two orders of magnitude lower than the above range (Mejia et al., 2007), and therefore, we did not expect that

Spatial variation in particle number size distributions

J. F. Mejía et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

they would significantly affect the size distribution.

4.1 Modal structures

As can be seen in Fig. 2, the modal structures and corresponding concentration levels were very similar to those reported from traffic and urban emissions (Morawska et al., 1999b). Particle number peaked at below 30 nm and a second peak was observed, or at least suggested by the results, in the range 50–100 nm, except at QUT, where the distribution was clearly unimodal. The results are consistent with the modal structures observed in petrol and diesel exhaust, whose respective NSDs are dominated by the ranges 20–60 and 20–120 nm (Maricq et al., 1999; Gouriou et al., 2004; Morawska et al., 1998a).

The first peak (<30 nm) predominates and is indicative of the predominance of petrol emissions in Brisbane, corroborating earlier findings (Holmes et al., 2005; Morawska et al., 1998b, 1999b), and also hint at the presence of diesel traffic (Morawska et al., 1998a). These particles belong to a size category known as nanoparticles (<50 nm), which are formed by primary carbonaceous emissions from petrol, and the supersaturation of semivolatile precursors due to the rapid cooling of both petrol and diesel traffic exhaust gas resulting in homogeneous nucleation (Bukowiecki et al., 2002; Imhof et al., 2005b; Kittelson et al., 2002). This has been shown to be a process that instantly (1–3 s) yields a high number of nanoparticles (Kittelson et al., 2000).

The second peak, in the range 50–100 nm, as already discussed, is indicative of diesel fuel. These particles consist predominantly of soot aggregates from incomplete combustion processes (Imhof et al., 2005a). Research from bench tests suggests that nanoparticles dominate the low engine load whereas the peak above 50 nm corresponds to high loads (Sturm et al., 2003). With the exception of QUT and Tora St, sampling took place close to traffic lights. During each scan, the lights changed, on average, ten times. As a result, engine loads varied regularly, being highest during acceleration.

Spatial variation in particle number size distributions

J. F. Mejía et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

The distributions at ANZ and Tora St are clearly dominated by the smaller particles, peaking at <25 nm. No peak was observed at ANZ site, and the data indicates that the peak is located below 15 nm. Modal diameters below this limit have been observed in urban air (e.g. Shi et al., 2001). Particle NSD has been observed to change with distance to the road, shifting to a larger diameter the further away downwind (Pirjola et al., 2006; Zhu et al., 2002). Research has found that the concentration of particles <50 nm decrease more rapidly than that of particles >100 nm and concluded that coagulation is more important than atmospheric dilution for UFPs whereas the reverse is true for larger particles (Zhu et al., 2002). Tora St and ANZ were very close to the road (<5 m) and therefore it is reasonable to conclude that the distributions observed at these two sites reflect their close proximity to traffic.

Of the other sites, only PAH was as close to the road. At this site, however, smaller particles are less dominant than at Tora St and ANZ. There is a greater flow of diesel vehicles around PAH, which partially explains the results. There were similarities but also differences in sampling conditions between PAH and the two sites above. As in Tora St, sampling took place directly above the road. The main difference was the traffic flow pattern, travelling continuously at 100–110 km hr⁻¹ at Tora St whereas at PAH sampling took place directly above a traffic light. Gouriou et al. (2004) and Imhoff et al. (2005a) observed that, for diesel fuel, particles >50 nm prevailed in stop-start traffic conditions. This can be explained by the higher engine loads associated with start-acceleration conditions near these lights, resulting in high emission rates of soot particles. At ANZ, sampling took place near a traffic light but unlike PAH it was at ground level. Research on the horizontal and vertical distribution of particles around buildings (Morawska et al., 1999a), however, found no association between height and total particle number concentration. The meteorological conditions were similar at the three sites, with average wind speeds ranging between 1.5 and 2.1 m/s. Therefore, although it is obvious that the observed distributions at these sites reflect their different environments, there are influences other than traffic or meteorological conditions yet to be identified.

Spatial variation in particle number size distributions

J. F. Mejía et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

The second mode observed at ANZ, unlike the other sites, occurs above 100 nm. During this campaign, there was an episode when the entire city was covered in smoke. Although this mode may explain the reduction in visibility, the episode was of very short duration, approximately one day. In addition, the spectrum looks quite different from that associated with bushfire activity (Morawska et al., 1999b). At this stage, the causes remain unclear.

BP site differs from the other sites in that the peak in the size distribution shifts to a larger diameter, around 50 nm. Free-flowing traffic at this site is dominated by diesel trucks, with average velocities of 70 km h⁻¹. The site was located at approximately 14 m above sea level, at the northern end of a bridge connecting the mainland to the seaport. Studies in different locations have consistently shown that particles associated with sea breeze have modal diameters in the range 100–200 nm (e.g. Bates et al., 2000; Eleftheriadis et al., 2006; Morawska et al., 1999b). Due to its location, sea breeze was expected to reach the site in all directions and therefore a peak in the above size range should be present. The absence of this peak therefore reflects the much stronger contribution of diesel traffic.

4.2 Cumulative distributions

Figure 3 shows that UFPs contribute between 82 and 89% of the total particle number and nanoparticles between 60% and 70%. This is consistent with the NSD observed in urban aerosols (Morawska et al., 1998a, b; Shi et al., 2001). At BP site, although the contribution of UFP is very consistent with the other sites, the contribution of nanoparticles drops to around 50%, reflecting the dominance of the larger ultrafine particles.

The K-S tests in Table 3 confirmed that all spectra had significantly different distributions. This supported the visible differences shown in Fig. 2. Despite the differences, the sites can be group into three major categories: (1) those with maximum concentrations in the 30 nm region (QUT, BSQ, PAH, Archerfield, Garden City and MGC Rd), (2) those with peak concentrations <25 nm (Tora St and ANZ), and (3), a site with peak concentrations at around 50 nm (BP). The modal structures in the first category

Spatial variation in particle number size distributions

J. F. Mejía et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

is characteristic of urban traffic emissions, particularly from petrol (Morawska et al., 1999b); the second category is clearly dominated by particles at the lower end of the size range, and it can be explained by the close proximity between the sites and the road; the spectrum in the third category is consistent with diesel emissions (Morawska et al., 1998a). Figure 1 shows that the sites corresponding to the first category are very widespread around the study area, those corresponding to the second category are very close to each other, surrounded by those in the first category and the site in the last category is clearly isolated from rest. Therefore, the interpretation of the data is influenced by the level of proximity between the sites.

The statistical analyses indicated that with only a few exceptions, wind direction or day of the week did not significantly affect the NSD. Based on what has already been discussed, it is reasonable to conclude that although the spectra are heterogeneously distributed probably due to local differences in the contribution of petrol vehicles. At BP, the contribution of nanoparticles becomes reduced and this can be attributed to the much larger percentage of diesel trucks at this site.

At the BSQ site, the distributions differed between weekdays and the weekend and this is reflected in the location of the corresponding peaks (Fig. 4). This site was located in the heart of the CBD. This difference is explained by the difference in fleet composition during these days. The CBD serves as a hub for public transport services, and 85% of buses operate on diesel fuel. Petrol traffic is more dominant during the weekend, resulting from a reduction in bus services, even though traffic flow rate is significantly reduced.

4.3 Number median diameters

In addition to the modal structure, the size distributions were analysed in terms of their NMDs (Fig. 5). Unlike the modal structures, some sites were found to have similar NMD values. Based on the statistical results and the average values, the sites were grouped into three major groups, and these groups were consistent with those obtained for the modal structures.

Spatial variation in particle number size distributions

J. F. Mejía et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Although some variations between sites were found within the groups, the magnitude of these differences was very small and not practically important. For instance, the NMDs consistently fall around or below the 40 nm region, which is a source signature of petrol traffic (Morawska et al., 1998b, 1999b). The only exception were BP site, where the average NMD is 59 nm, reflecting the predominance of diesel trucks (Morawska et al., 1998a).

To deepen the analysis, the GSD were also compared and there was little variability among the sites, with average values falling around 1.96, although it was visibly higher at BP (2.46). In order to shed more light into the shape of the distributions, the correlation between NMD and GSD was analysed and although it was significant at the 99% confidence level ($p < 0.01$), it was rather weak ($r = -0.14$). Therefore, the effect of NMD on the width of the distribution may not be important in practice.

5 Summary and conclusions

To summarise the findings from this study, the data were collected during different campaigns of varying durations spanning an 11-year period. The results indicated that there were spatial and temporal variations in the modal structures and therefore that particle number size was heterogeneously distributed in Brisbane. UFPs and nanoparticles contributed, respectively, to around 82–89% and 60–70% of the particle number. Although the contribution of UFPs at the BP site was similar to the other sites, the contribution of nanoparticles dropped to 50%. Particle number peaked at around or below 30 nm and NMD in the 40 nm region. Although some differences were also found for the NMD, there were four sites that had similar NMD values. Where statistically significant differences were found, in most cases, their magnitudes were very small and therefore not important in practice. Although the locations of the peaks varied between sites, in most cases half of the particle number was within approximately the same size range. Based on these results, the sites were divided into three major groups: (1) urban sites dominated by petrol traffic emissions, (2) sites dominated by the smaller

Spatial variation in particle number size distributions

J. F. Mejía et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

particles probably caused by the proximity between the measurement points and the road, and (3) a site clearly dominated by diesel traffic. The NSDs were clearly unaffected by differences in sampling periods. The locations of the peaks and NMD values consistently indicate that petrol vehicles are the dominant source, and therefore the results from the statistical tests hint at differences in traffic composition, as expected. At this stage, however, no association between traffic composition and size distribution could be established due to the limited traffic information available for this study. Future investigations should benefit by including traffic composition data.

Acknowledgements. The authors wish to express their appreciation to R. Jayaratne, G. Johnson and N. Holmes, from the International Laboratory of Air Quality and Health (ILAQH) and M. Jamriska (formerly from ILAQH and currently from the Australian Defence Academy), for supplying their data for the purposes of this project. Funding from the Australian Research Council is also gratefully acknowledged.

References

Abu-el-zeet, Z. H., Becerra, V. M., and Roberts, P. D.: Combined bias and outlier identification in dynamic data reconciliation, *Comput. Chem. Eng.*, 26, 921–935, 2002.

Barnett, V. and Lewis, T.: *Outliers in statistical data*, Chichester, Wiley, 1994.

Bates, T. S., Quinn, P. K., Covert, D. S., Coffman, D. J., Johnson, J. E., and Wiedensohler, A.: Aerosol physical properties and processes in the lower marine boundary layer: a comparison of shipboard sub micron data from ACE 1 and ACE 2, *Tellus B*, 52, 258–272, 2000.

Bukowiecki, N., Dommen, J., Prevot, A. S. H., Richter, R., Weingartner, E., and Baltensperger, U.: A mobile pollutant measurement laboratory—measuring gas phase and aerosol ambient concentrations with high spatial and temporal resolution, *Atmos. Environ.*, 36, 5569–5579, 2002.

Bureau of Meteorology: *The Climate of Brisbane*, Canberra, 1983.

Buzorius, G., Hameri, K., Pekkanen, J., and Kulmala, M.: Spatial variation of aerosol number concentration in Helsinki city, *Atmos. Environ.*, 33, 553–565, 1999.

CONCAWE: *Measurement of the Number and Mass Weighted Size Distributions of Exhaust*

Spatial variation in particle number size distributions

J. F. Mejía et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Particles Emitted from European Heavy Duty Engines (Report No. 01/51), Brussels, CON-CAWE, 2001.

Eleftheriadis, K., Colbeck, I., Housiadas, C., Lazaridis, M., Mihalopoulos, N., Mitsakou, C., Smolik, J., and Zdimal, V.: Size distribution, composition and origin of the submicron aerosol in the marine boundary layer during the eastern Mediterranean "SUB-AERO" experiment, Atmos. Environ., 40, 6245–6260, 2006.

Flocas, H. A., Assimakopoulos, V. D., and Helmis, C. G.: An experimental study of aerosol distribution over a Mediterranean urban area, Sci. Total Environ., 367, 872–887, 2006.

Gibbons, J. D. and Chakraborti, S.: Nonparametric Statistical Inference, New York, Marcel Dekker Inc., p. 583, 2003.

Gouriou, F., Morin, J.-P., and Weill, M.-E.: On-road measurements of particle number concentrations and size distributions in urban and tunnel environments, Atmos. Environ., 38, 2831–2840, 2004.

Harrison, R. M., Jones, M., and Collins, G.: Measurements of the physical properties of particles in the urban atmosphere, Atmos. Environ., 33, 309–321, 1999.

Heitinbrink, W. A., Baron, P., and Willeke, K.: Coincidence in time-of-flight aerosol spectrometers: phantom particle creation, Aerosol Sci. Tech., 14, 112–126, 1990.

Hitchins, J., Morawska, L., Wolff, R., and Gilbert, D.: Concentrations of submicrometre particles from vehicle emissions near a major road, Atmos. Environ., 34, 51–59, 2000.

Holmes, N. S., Morawska, L., Mengersen, K., and Jayaratne, E. R.: Spatial distribution of submicrometre particles and CO in an urban microscale environment, Atmos. Environ., 39, 3977–3988, 2005.

Hong, Y. C., Lee, J. T., Kim, H., Ha, E. H., Schwartz, J., and Christiani, D. C.: Effects of air pollutants on acute stroke mortality, Environ. Health Persp., 110, 187–191, 2002.

Hussein, T., Hameri, K., Aalto, P. P., Paatero, P., and Kulmala, M.: Modal structure and spatial-temporal variations of urban and suburban aerosols in Helsinki-Finland, Atmos. Environ., 39, 1655–1668, 2005.

Hussein, T., Puustinen, A., Aalto, P., Makela, T., Hameri, K., and Kulmala, M.: Urban aerosol number size distributions, Atmos. Chem. Phys., 4, 391–411, 2004, <http://www.atmos-chem-phys.net/4/391/2004/>.

Imhof, D., Weingartner, E., Ordonez, C., Gehrig, R., Hill, M., Buchmann, B., and Baltensperger, U.: Real-World Emission Factors of Fine and Ultrafine Aerosol Particles for Different Traffic Situations in Switzerland, Environ. Sci. Technol., 39, 8341–8350, 2005a.

ACPD

7, 17147–17177, 2007

Spatial variation in particle number size distributions

J. F. Mejía et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

EGU

Imhof, D., Weingartner, E., Vogt, U., Dreiseidler, A., Rosenbohm, E., Scheer, V., Vogt, R., Nielsen, O. J., Kurtenbach, R., and Corsmeier, U.: Vertical distribution of aerosol particles and NO_x close to a motorway, *Atmos. Environ.*, 39, 5710–5721, 2005b.

Jaenicke, R.: Tropospheric aerosols, in: *Aerosol-Cloud-Climate Interactions*, edited by: Hobbs, P. V., London, Academic Press, 1993.

Kittelson, D., Watts, W., and Johnson, J.: *Diesel Aerosol Sampling Methodology – CRC E-43*, Final report, Coordinating Research Council, 2002.

Kittelson, D. B., Johnson, J., Watts, W., Wei, Q., Drayton, M., Paulsen, D., and Bukowiecki, N.: Diesel sampling in the atmosphere, *SAE Technical Paper Series No. 2000-01-2212*, 2000.

Maricq, M. M., Podsiadlik, D. H., and Chase, R. E.: Examination of the Size-Resolved and Transient Nature of Motor Vehicle Particle Emissions, *Environ. Sci. Technol.*, 33, 1618–1626, 1999.

Mejia, J. F., Wraith, D., Mengersen, K., and Morawska, L.: Trends in size classified particle number concentration in subtropical Brisbane, Australia, based on a 5 year study, *Atmos. Environ.*, 41, 1064–1079, 2007.

Morawska, L., Bofinger, N., Kocis, L., and Nwankwoala, A.: Submicrometer and supermicrometer particles from diesel vehicle emissions, *Environ. Sci. Technol.*, 32, 2033–2042, 1998a.

Morawska, L., Jayaratne, E. R., Mengersen, K., Jamriska, M., and Thomas, S.: Differences in airborne particle and gaseous concentrations in urban air between weekdays and weekends, *Atmos. Environ.*, 36, 4375–4383, 2002.

Morawska, L., Thomas, S., Bofinger, N., Wainwright, D., and Neale, D.: Comprehensive characterization of aerosols in a subtropical urban atmosphere: particle size distribution and correlation with gaseous pollutants, *Atmos. Environ.*, 32, 2467–2478, 1998b.

Morawska, L., Thomas, S., Gilbert, D., Greenaway, C., and Rijnders, E.: A study of the horizontal and vertical profile of submicrometer particles in relation to a busy road, *Atmos. Environ.*, 33, 1261–1274, 1999a.

Morawska, L., Thomas, S., Jamriska, M., and Johnson, G.: The modality of particle size distributions of environmental aerosols, *Atmos. Environ.*, 33, 4401–4411, 1999b.

Pirjola, L., Paasonen, P., Pfeiffer, D., Hussein, T., Hämeri, K., Koskentalo, T., Virtanen, A., Rönkkö, T., Kesniken, J., Pakkanen, T. A., and Hillamo, R.: Dispersion of particles and trace gases nearby a city highway: mobile laboratory measurements in Finland, *Atmos. Environ.*, 40, 867–879, 2006.

Salma, I., Dal Maso, M., Kulmala, M., and Zaray, G.: Modal characteristics of particulate matter

Spatial variation in particle number size distributions

J. F. Mejía et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

- in urban atmospheric aerosols, *Microchem. J.*, 73, 19–26, 2002.
- Seaton, A., Godden, D., MacNee, W., and Donaldson, K.: Particulate air pollution and acute health effects, *Lancet*, 345, 176–178, 1995.
- Seinfeld, J. H. and Pandis, S. N.: *Atmospheric Chemistry and Physics: From air pollution to climate change*, John Wiley & Sons Inc., New York, 1998.
- Shi, J. P., Evans, D. E., Khan, A. A., and Harrison, R. M.: Sources and concentration of nanoparticles (<10 nm diameter) in the urban atmosphere, *Atmos. Environ.*, 35, 1193–1202, 2001.
- Sturm, P. J., Hausberger, S., Engler, D., Bacher, M., Lechner, B., and Rodler, J.: PM size distribution measurements in engine exhaust in context to road tunnel and ambient air measurements, *Fifth international technion symposium*, Vienna, Austria, 2003.
- Virtanen, A. K. K., Ristimäki, J. M., Vaaraslahti, K. M., and Keskinen, J.: Effect of Engine Load on Diesel Soot Particles, *Environ. Sci. Technol.*, 38, 2551–2556, 2004.
- Wåhlin, P., Palmgren, F., and Van Dingenen, R.: Experimental studies of ultrafine particles in streets and the relationship to traffic, *Atmos. Environ.*, 35, 63–69, 2001.
- Yu, T. C., Teh, H. W., Jacques, P. A., Sioutas, C., and Froines, J. R.: Quality control of semi-continuous mobility size-fractionated particle number concentration data, *Atmos. Environ.*, 38, 3341–3348, 2004.
- Zhu, Y., Hinds, W. C., Kim, S., and Sioutas, C.: Concentration and size distribution of ultrafine particles near a major highway, *J. Air Waste Manage.*, 52, p. 1032, 2002.

Spatial variation in particle number size distributions

J. F. Mejía et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

**Spatial variation in
particle number size
distributions**

J. F. Mejía et al.

Table 1. Summary of the monitoring sites.

Site	# Type of Environment	Travel Speed (km h ⁻¹)	Monitoring Period	*Recorded Observations	Scan Time (s)	Data Retention (%)	Approx. Distance to CBD
QUT AMRS	Urban-Freeway	70–90	1 June 1995–7 Nov 2000	24 026	90	83.3	Within CBD
Brisbane Square (BSQ)	Urban-Commercial	50–60	18–24 Jan 2004	6 458	180	99.5	Within CBD
PAH	Commercial-Suburban	60–70	30 July–23 Oct 1999	8 390	300	99.9	4 km S
Tora St	Freeway	100–120	16 Sep 1998–3 June 1999	8626	300	89.2	10 km S
Archerfield	Industrial-Commercial	60–70	17–24 June 2002	1 802	300	100.0	11 km S
ANZ	Commercial-Suburban	60–70	1–8 July 2002	1 917	300	98.7	11 km S
Garden City	Commercial	60–70	24 June–1 July 2002	1 561	300	100.0	11 km S
Mt Gravatt-Capalaba Rd (MGC Rd)	Suburban	60–70	6–17 June 2002	1 733	300	100.0	11 km S
Port of Brisbane (BP)	Marine-Industrial	60–70	12 May–9 June 2006	5 904	300	99.6	16 km NE

With the exception of the freeway type, which is assumed to be continuous non-stop driving mode, traffic is assumed to travel in stop-start mode.

*Submicrometre particles only.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

**Spatial variation in
particle number size
distributions**

J. F. Mejía et al.

Table 2. Recorded temperatures (°C) in each site.

Site	¹ Minimum	Maximum	Average
QUT AMRS	0.0	40.0	21.0
Brisbane Square (BSQ)	8.0	35.5	26.4
PAH	5.9	27.7	16.5
Tora St	9.3	35.2	21.9
Archerfield	0.4	24.9	14.9
ANZ	5.7	25.2	14.3
Garden City	6.7	23.7	14.0
Mt Gravatt-Capalaba Rd (MGC Rd)	6.9	30.7	16.6
Port of Brisbane (BP)	9.0	25.1	17.2

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

I◀

▶I

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

**Spatial variation in
particle number size
distributions**

J. F. Mejía et al.

Table 3. Kolmogorov-Smirnov test results on average spectra.

Site	ANZ	Archerfield	BP	BSQ	Garden City	MGC Rd	PAH	QUT
Archerfield	0.122							
BP	0.171	0.098						
BSQ	0.146	0.049	0.073					
Garden City	0.122	0.024	0.098	0.024				
MGC Rd	0.122	0.024	0.073	0.024	0.024			
PAH	0.098	0.024	0.098	0.049	0.024	0.049		
QUT	0.143	0.049	0.073	0.024	0.049	0.024	0.049	
Tora St	0.049	0.122	0.171	0.122	0.122	0.122	0.098	0.122

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Spatial variation in particle number size distributions

J. F. Mejía et al.

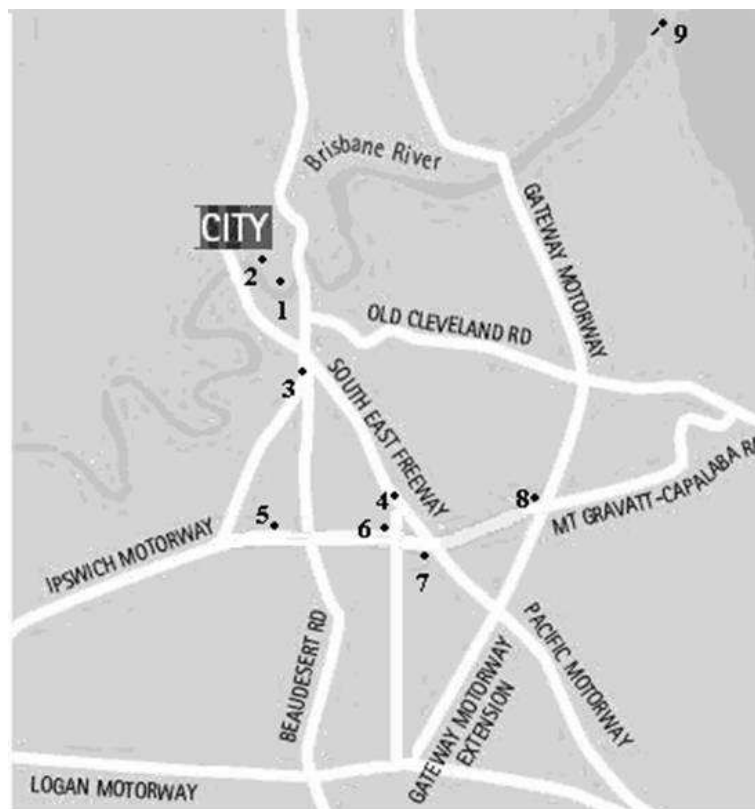


Fig. 1. Schematic representation (not to scale) of the sampling point locations: (1) QUT, (2) Brisbane Square, (3) Princess Alexandra Hospital, (4) Tora St, (5) Archerfield, (6) ANZ Sports Complex, (7) Garden City, (8) Mt Gravatt-Capalaba Rd, and (9) Brisbane Port.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Spatial variation in particle number size distributions

J. F. Mejía et al.

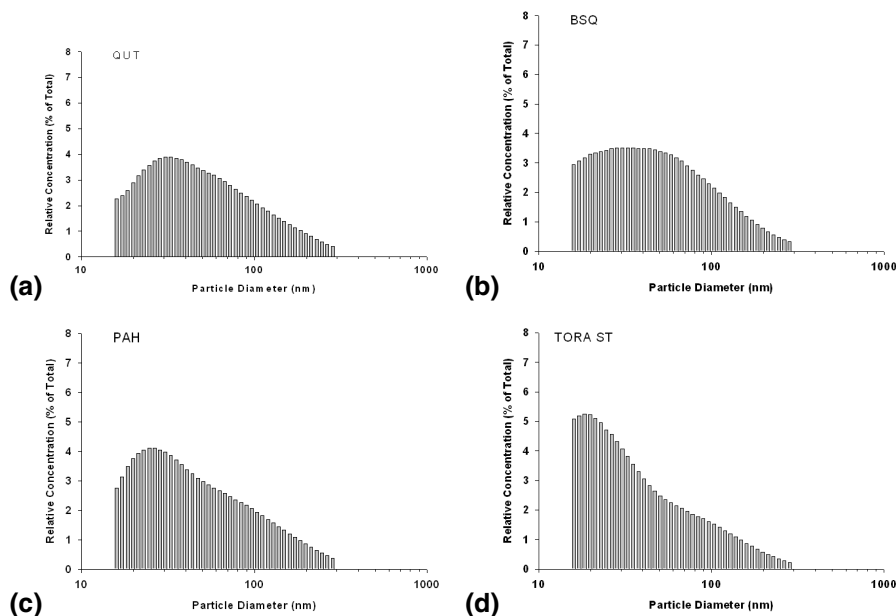


Fig. 2. Average particle NSD spectra in the range 15–300 nm of data collected at nine different locations in Brisbane, Australia.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[I◀](#)[▶I](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

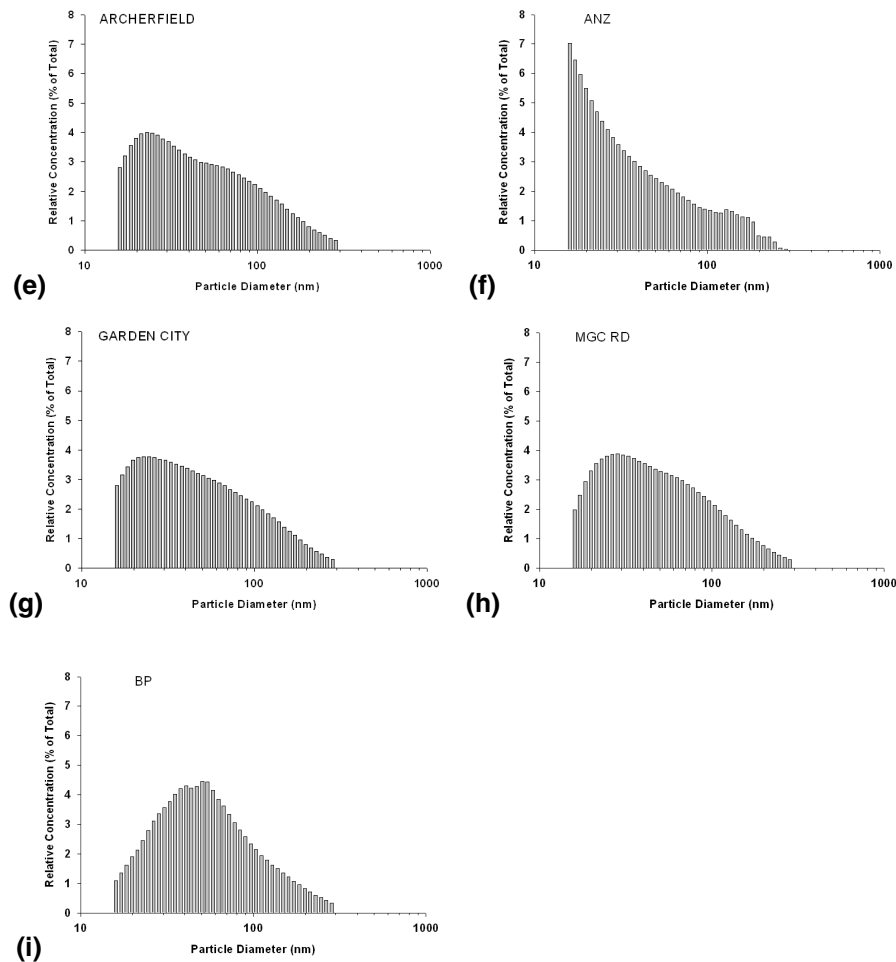


Fig. 2. Continued.

Spatial variation in particle number size distributions

J. F. Mejía et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Spatial variation in particle number size distributions

J. F. Mejía et al.

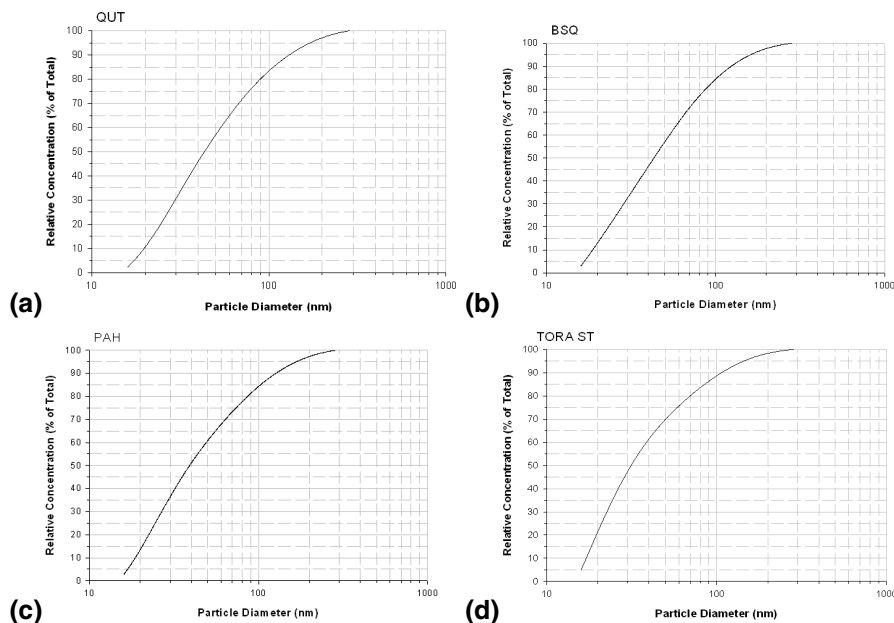


Fig. 3. Cumulative particle NSD in the range 15–300 nm obtained from the average modal structure obtained at the nine selected sites.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[I◀](#)[▶I](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Spatial variation in particle number size distributions

J. F. Mejía et al.

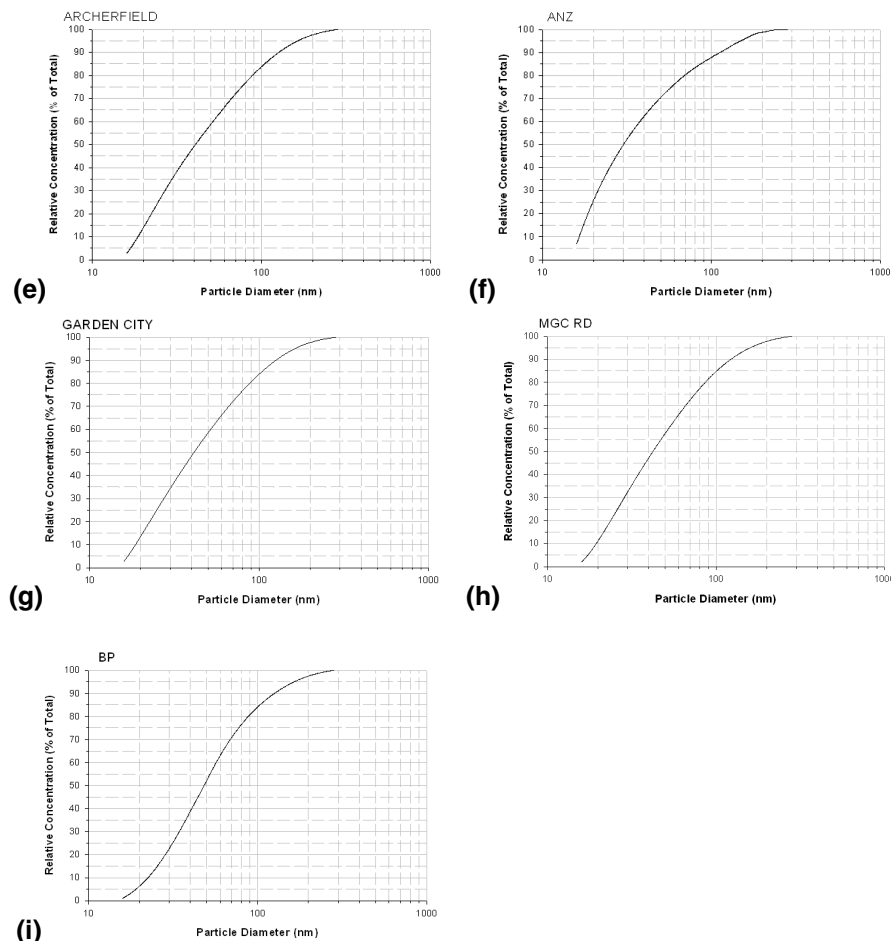


Fig. 3. Continued.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

Spatial variation in particle number size distributions

J. F. Mejía et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

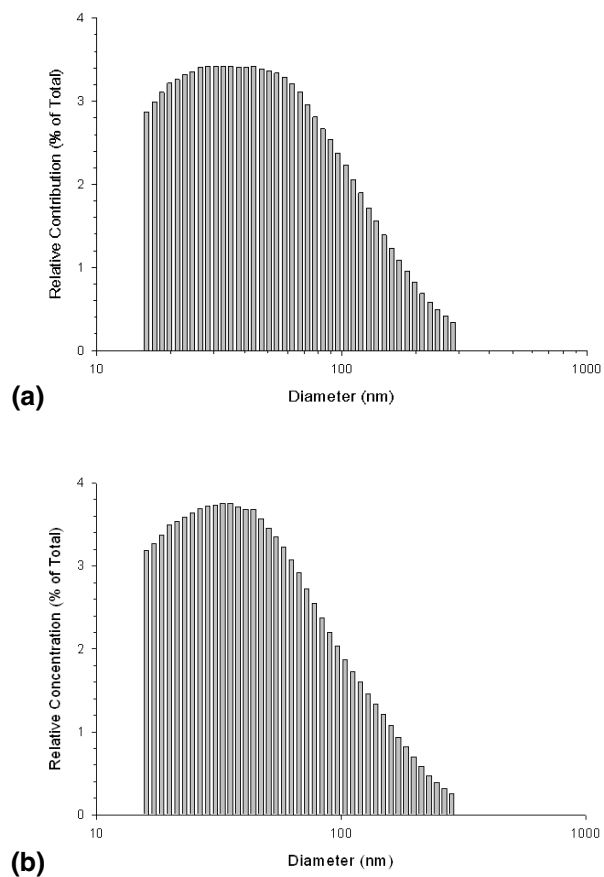


Fig. 4. Observed differences in NSD spectra obtained at BSQ during (a) weekdays and (b) the weekend.

Spatial variation in particle number size distributions

J. F. Mejía et al.

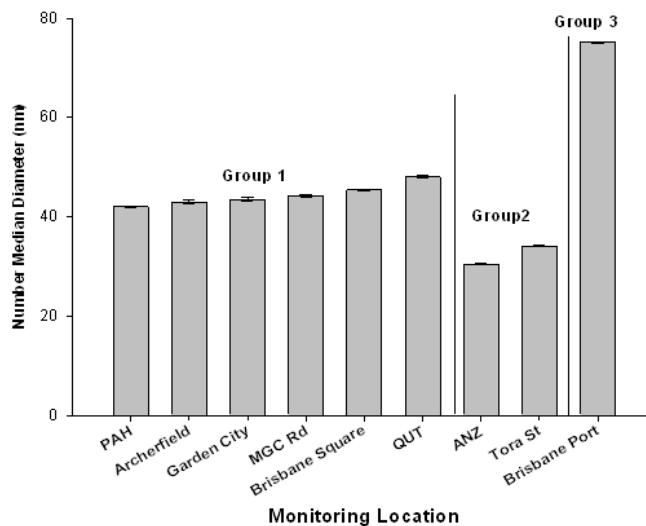


Fig. 5. Average NMD per site. The error bars represent the corresponding standard deviations. The continuous lines are used to group the sites into three major categories.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)