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Comment

Interactive comment on “Spatial variation in particle number size distributions in a largemetropolitan area” by J. F. Mejía et al.

J. F. Mejía et al.

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Reviewer #1:

General Comments:

The paper describes an important topic of spatial and temporal variation of number size distributions (NSD) in a city. The text is easy to follow, quite compact and well written. One problem may be that now the text in 3 Results is much repeated in 4 Discussion. So, combining these two could shorten and clarify the text. Literature is widely cited and the technical details are properly explained. The conclusions are interesting and seem to have a strong basis on the measurement data, although some more details could be explained. Especially I would like to see a figure or a sentence clarifying how is the difference/similarity between average winter and average summer

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time NSDs at sites where this data is available. Of interest are also the average total number concentration levels at each site, which information could be easily included for instance in one of the tables or in Figure 2. The average data from many sites is similar, which suggests that the temporal variation of average NDSs has been small in the eleven year period. However, the text about this topic is quite scattered in the present paper, and could be focussed better. At some points the text is difficult to understand.

Response to General Comments:

Given the length of the comments, I found it convenient to divide it into several parts. So I will address each issue in the same order as it appeared in the comments:

1. One problem may be that now the text in 3 Results is much repeated in 4 Discussion. So combining these two could shorten and clarify the text.

This comment is rather vague, so an effort was made to identify where the repetitions occur. After a detailed reading, we agree that the description of results in page 17157 lines 8-12 (page 11 line 23 to page 12 line 3 in original text) where it describes the three major groups of spectra, are very much the same in the discussion in page 17161 lines 26-28 (page 17 lines 2-6). Therefore the lines were eliminated from the results section (page 17157) and the new text reads:

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. 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.

2. The conclusions are interesting and seem to have a strong basis on the measurement data, although some more details could be explained. Especially I would like to see a figure or a sentence clarifying how is the difference/similarity between average

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winter and average summer time NSDs at sites where this data is available.

The comments on the differences and similarities are based on the statistical tests. To explain it more clearly, a section was added to the discussion explaining why there is no seasonality in Brisbane. The test reads as follows (page 17163, line 13):

4.4 Effect of Seasonality: The interpretation of the results suggests that, with the exception of BP site, the distributions were unaffected by the differences in sampling period. A time series investigation (Mejia et al., 2007) found that, unlike in temperate latitudes, particle size distribution in Brisbane is unaffected by season of the year. This conclusion was based on seasonal analysis results on particle number and NMD, which showed no annual periodic behaviour. The lack of seasonality in Brisbane can be explained by the very small variations in meteorological conditions, for example air temperature, which in this study had a range of 10oC between the minimum and maximum averages (Table 2). There were obvious differences in particle number (Table 2), but these can be interpreted as resulting from varying distance to the road, traffic levels and composition rather than caused by seasonal differences.

3. Of interest are also the average total number concentration levels at each site, which information could be easily included for instance in one of the tables or in Figure 2.

Minimum, maximum and average total concentrations were added to Table 2. Also, reference to the concentrations were made in Section 3.3 (see answer to next comment) and in the discussion (see answer to previous comment).

4. The average data from many sites is similar, which suggests that the temporal variation of average NDSs has been small in the eleven year period. However, the text about this topic is quite scattered in the present paper, and could be focussed better.

Please refer to section 3.3, where it mentions that the differences between campaigns were very small. To make it clear, the ranges of values were added to this section (page 17156, lines 13-16) and the text reads as follows:

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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 fell respectively, around 30 and 40nm, with the exception of BP site where the corresponding values were around 50 and 60 nm. Similarly the ranges for the contribution of ultrafine particles and nanoparticles to the total concentration were very narrow (around 10% between their corresponding minima and maxima). The only exception was BP site but this cannot be explained by seasonal differences, particularly of temperature, as the averages range between 14.0oC and 26.4oC (Table 2). As Table 2 shows, although there are obvious differences in particle concentration levels (ranging from 7.2 to 45.6 x 10³ cm⁻³), the range of temperatures is very narrow, with average values ranging between 14.0oC and 26.5oC. Therefore, it is very clear that the sampling periods had no effect on the distributions and particle concentration levels.

5. The text is easy to follow, quite compact and well written … At some points the text is difficult to understand.

There is a contradiction at the beginning of the comment and at the end. The text is either well-written or difficult to follow. Also, it is very vague. Without indicating what points were difficult to understand, it leaves the author making guesses. The paper was proofread prior to submission and the initial nominated reviewers didnt have any difficulties reading the text.

Specific Comments

1. Abstract: Lines 14-15, give size for UFP

The size for UFP has been added to the text, the line reads: Ultrafine particles (UFPs (< 100 nm))

2. Introduction: Page 17149, line 7 - is it really mass of UFP here

To make the text clear, it has been changed as follows:

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it has been suggested that the observed effects could be better explained by the number of the ultrafine fraction (< 100 nm) rather than the mass of the larger particles.

3. Page 17149, line 28: collected by which instruments?

The current instruments employed for the measurement of the number size distribution are TSI scanning mobility particle sizer (SMPS) systems. Generally, unless a new instrument is presented, instruments used for the collection of airborne samples is not mentioned. The paragraph focuses on the need to measure the size distribution and to make the objective clearer - collected samples - was changed to -distribution. In other words, the sentence was modified as follows:

the analysis of particle size distribution can be useful in the interpretation of the properties of the distribution.

4. Methodology: Page 17151, line 6 - background emissions?

To make it clear, background was changed to non-freeway.

5. Give percentage of outliers at each site.

In section 2.4, page 17155, lines 4-5, where it explains that outliers were identified and removed. And in Table 1, it provides the information on the level of data retention per site.

6. Results: Page 17156, line 8 - should be less than 300 nm (not submicrometre)

A definition for submicrometre particles as used in the paper has been added and the sentence:

Figure 2 shows the average modal structure of the submicrometre (referred in this paper, unless otherwise indicated to particles < 300 nm) particle NSD obtained from each site.

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7. Page 17156, line 13 - likely all NSDs have at least two modes, but partly masked

This is a very fair comment and the sentence has been modified as follows:

...the NSDs were likely to have at least two peaks, but partly masked.

8. Pages 17157-58 - first the differences were significant, but then very small?

To clarify this confusion, what it meant was that although the statistical tests indicated that the differences were statistically significant, the maximum range of these differences was only 5 nm, which is very small in practice. Therefore, the first sentence in section 3.2 has been modified as follows (page 17157, line 25):

ANOVA tests applied to NMD found statistically significant differences between sites.

9. Discussion: Page 17160, lines 6-9 - note that some other studies have realized that the importance of atmospheric dilution is the main process of UFP too.

This is a very important observation and therefore a sentence has been added to line

9. The amended text reads as follows (p. 17160, line 6):

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., 2002b). Other investigations, on the other hand, have found that dilution is as important as coagulation in changing the concentration of UFPs with increasing distance to the road (eg. Zhu et al., 2002a).

Since the cited works refer to the same first author but different investigations, all previous references to Zhu et al. 2002 has been changed to Zhu et al. 2002b, and the corresponding works in the reference list are:

Zhu, Y., Hinds, W. C., Kim, S., Shen, S. & Sioutas, C. (2002a) Study of ultrafine particles near a major highway with heavy-duty diesel traffic. *Atmospheric Environment*, 36, 4323-4335. Zhu, Y., Hinds, W. C., Kim, S. & Sioutas, C. (2002b) Concentration and

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size distribution of ultrafine particles near a major highway. Journal of the Air & Waste Management Association, 52, 1032.

10. Page 17161, line 3, which mode?

To make it clear, the sentence has been modified as follows:

A peak above 100 nm is observed at ANZ site although its concentration is close to the lowest in the ultrafine range.

11. Page 1761: explain see breeze in more detail. One could expect sea-salt particles there.

Modification has been made to the text to explain the NSD of sea salt:

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, travelling with average velocities of 70 km h⁻¹. The smaller peak at 40 nm is consistent with the peak observed in sea breeze (Morawska et al., 1999) probably resulting from homogeneous nucleation in marine boundary layer (Hoppel et al., 1990). The site was located at approximately 14 m above sea level, at the northern end of a bridge connecting the mainland to the seaport and therefore it is reasonable to interpret this peak as being associated with sea breeze. However, based on the NSD on marine aerosols (eg. Bates et al., 2000; Eleftheriadis et al., 2006; Morawska et al., 1999) a small peak in the range 100-200 nm was expected. The absence of this peak at the site therefore reflects the much stronger contribution of diesel traffic.

Note that a new reference was inserted and the details are:

Hoppel, W. A., Larson, R. & Vietti, M. A. (1990) Aerosol size distributions and optical boundaries found in the marine boundary layer over the Atlantic Ocean. Journal of Geophysical Research 95 D4, pp. 3659-3686. Journal of Geophysical Research, 95, 3659-3686.

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12. Page 17162, lines 11-13 - unclear

A typing error was detected. The word "although" was eliminated from the sentence. The corrected text reads:

Based on what has already been discussed, it is reasonable to conclude that the spectra are heterogeneously distributed probably due to local differences in the contribution of petrol vehicles.

13. Page 17163: likely the GSDs are more useful for single modes than for the sum of several modes

We fully agree with the reviewer's comments. However, because the dominance of one mode in the NSD was clearly visible, to the point of masking the second mode, it was considered useful to analyse the GSDs for the entire range, in the same way we calculated the NMD.

14. Summary, page 17163 Lines 2-4 - unclear

The lines were proofread and none of the previous reviewers had a problem with these lines. However, to make it clearer, the sentence has been somewhat modified:

The results indicated that there were statistically significant differences in the modal structures and therefore they showed that particle number size was heterogeneously distributed in Brisbane.

15. Page 17164, line 2: better to say: average NSDs, because there surely are differences between day and night.

This is an important observation. It was earlier mentioned in page 17157, lines 16-24 that there were no significant differences between data subsets for each site (by wind direction or day of the week). Nonetheless, the above comment is quite valid and therefore the above change has been made in p 17164, line 2:

The average NSDs were clearly

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16. The different character of the BP site is not mentioned here

The summary mentions the difference of the distribution at BP site, in lines 19-20. But to expand it a little further, the sentence has been modified as follows:

Although the contribution of UFPs at the BP site, was similar to the other sites, the contribution of nanoparticles decreased to 50%, reflecting its different environment.

17a) Table 1: Recorded observations, but for 1 min or for 1 hour or what?

The footnote - *Submicrometre particles only - has been changed to - *Based on 5-minute averages of submicrometre particles.

17b) Table 1: Explain CBD

A superscript has been added to CBD (**CBD) and a footnote have been added. The footnote reads **Central Business District of Brisbane.

17c) Table 2: For Minimum there is a superscript “1”?”?

This was a typing error that has been corrected and the superscript has therefore been deleted.

17d) Table 3: one station is missing! Which numbers mean high similarity?

The table is complete. All possible K-S results (36) have been double checked to make sure that no station is missing. The note at the bottom of the table has been replaced with: “Differences are significant at the 95% confidence level if K-S value > 0.020.

18. Figure 1: Give the scale for the map

A scale has been added to the figure and the title has been modified as follows:

Figure 1: Schematic representation (approximate scale: 5.4 mm = 1 km) 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-

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Capalaba Rd, and (9) Brisbane Port.

Technical Corrections

1. Page 17155, line 11, below 300 nm

The correction has been made to: < 300 nm

2. Page 17158, line 2: NMD instead of particles

The above correction has been made.

Anonymous Referee # 2

General Comments:

The paper presents interesting and important data on the spatial variation of number size distributions of particles

1. Methodology: OK. Comment: at some sites, quite short monitoring periods (one week) at others: 5 years. Seasons are different. The paper concludes that this has no influence on the statistics. But meteorological conditions are different between seasons: explain why no difference occurred.

A section has been added to the discussion (see reply to referee # 1).

2. Results and Figures: Fig 2. Text says - bimodal distributions can be found... It is not easy to recognize the bimodal function in these figures.

In page 17156, lines 11-13 it says that - at most sites, there were two peaks or modes: one occurring below 30 nm, and a second peak or protrusion suggesting its presence. It wasn't meant to be categorical of bimodality, but only that it was suspected.

In reply to the comment by reviewer # 1, the text has been changed to - As shown by these results, the NSDs were likely to have at least two peaks, but partly masked: one occurring below...

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3. Conclusions: a more concise comment on potential sources and the occurrence of size distribution is needed (which source(s) add to which size).

In the discussion section the paper explains why the NSD indicate a predominance of petrol traffic emissions, and the effect of proximity to the road. Based on the discussion, in page 17164, lines 3-5 read:

The locations of the peaks and NMD values consistently indicate that petrol vehicles are the dominant source, and therefore...

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 17147, 2007.

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