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Atmospheric Chemistry and Physics Discussions

Interactive comment on "An investigation of nucleation events in a coastal urban environment in the Southern Hemisphere" *by* J. F. Mejía and L. Morawska

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General Comments:

8220;The manuscript describes an analysis of a particle size distribution data set collected at an industrial site in Brisbane Australia aiming to investigate nucleation event frequencies in the area. The subject of this work is interesting, as more long-term data of secondary particle formation are needed from different types of environments. However, I think that the data set and, more importantly, its analysis is not appropriate to draw any conclusions of regional scale particle formation. Because of this, I think that the material presented in this work is not sufficient for a full paper, and its presentation is somewhat unclear in many places. I therefore think that this manuscript is not ready

to be published in ACP without compete re-thinking of presentation of this data set and possible re-submission8221;.

Response to General Comments:

The data was carefully assessed and analysed to investigate sources of secondary particles in the study area and tables 1 and 2 in the original manuscript provide information on the atmospheric conditions during nucleation events., the reviewer8217;s comment that more material is needed was addressed conscientiously, although a more specific comment would have aided us to correct those parts of the manuscript that the reviewer thought needed modification. Therefore, the corrections to the text we made might not be consistent with what the reviewer might have had in mind. The following corrections were made in response to these general comments (original text in normal font format, modified text in italics):

1. One major modification was to add some sample plots of times when new particle formation was observed (see specific comment 3).

2. A new line was added to page 2207, lines 13-15, after

8221; The presence of a high particle surface area prevents nucleation due to diffusion of small particles and condensed material to the surface of larger particles (e.g. Friedlander et al., 1991). As a result, nucleation events were rare and of very short duration8221;.

The modified text now reads:

8220;The presence of a high particle surface area prevents nucleation due to diffusion of small particles and condensed material to the surface of larger particles (Friedlander et al., 1991). This led us to hypothesise that air masses associated with nucleation events mixed quickly with emissions from the road. As a result, nucleation events were rare and of very short duration8221;.

In a like manner, the conclusion that 8220;nucleation events were associated with

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cleaner air masses and long-range transport8221; (pages 2208 line 28 8211; 2209 line 1) was replaced with:

8220;nucleation events were associated with cleaner air masses of local origin that mixed quickly with road emissions after the events, causing them to be short-lived 8221;.

3. The corresponding correction was made in the abstract. (page 2196, lines 13-16: 8220;The events were associated with sea breeze and long-range transport. Roadside emissions, in contrast, did not contribute to nucleation, probably due to the predominance of particles in the range 508211;100 nm associated with these emissions8221;

The corresponding correction now reads

8220;Roadside emissions, in contrast, did not contribute to nucleation, probably due to the predominance of particles in the range 50-100 nm associated with these emissions. The events were associated with clean air masses of local origin that quickly mixed with roadside emissions, causing these events to be of very short duration8221;.

4. The two paragraphs covering page 2205 (line 29) to page 2206 (line 25) have been modified, adding a paragraph in between. The new text reads:

8220;Global radiation has been identified to influence new particle production (Boy Kulmala, 2002; O'Dowd et al., 1999). Solar radiation levels are generally higher in summer and lower in winter and as a result American and European studies have found nucleation events to be more frequent in summer (e.g. Qian et al., 2007). Conversely, other studies have found that nucleation events are less frequent winter than in spring or autumn (e.g. Stanier et al., 2004) and sometimes they are even absent during the winter (Wehner Wiedensohler, 2003). In contrast, a South Korean study found that nucleation events occurred more frequently in winter (Lee et al., 2008). The difference between the South Korean study and the European and American studies is that the South Korean study was done in a coastal environment whereas those in Europe and

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the USA were conducted in urban areas.

In the present study, although it took place in a coastal area, nucleation had a higher frequency in summer than in winter, which is more consistent with the urban studies above than with the coastal study in South Korea. Although this may reflect the high traffic activity at the site, thereby suggesting photochemical formation, the fact that none of them took place when the wind blew from the road indicates that they are not associated with traffic. These events were rarer in spring and absent in the autumn campaigns.

The statistical test showed that the difference in solar radiation levels during these events was almost the same between summer and winter. Furthermore, many of these events were observed at much lower radiation levels than those observed in East St. Louis (Qian et al., 2007). In that study, nucleation was observed when the median solar radiation intensity was 680 W m-2 and no nucleation events were observed at below 450 W m-2 (Qian et al., 2007). In the present investigation, nucleation events were observed even when solar radiation intensity was as low as 46 W m-2. This compares well with the conditions observed in coastal environments (e.g. Lee et al., 2008). The occurrence of new particle production depends not only on the presence of intense solar radiation but also on the properties of the present air mass (Wehner Wiedensohler, 2003). Therefore, although the importance of solar radiation upon new particle formation cannot be dismissed, there are other forces influencing the process, for example, the type of environment.8221;

5. A clarification needs to be made that the focus of the investigation to assess the contribution of traffic emissions to secondary particle formation rather than analyse regional scale particle formation. To clarify this, the text was modified (last paragraph in page 2197, lines 21-26). The original text was:

8220;This paper aims to analyse the frequency of and the atmospheric conditions favourable for nucleation events at coastal urban location in Brisbane, Australia. Mon-

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itoring was conducted during four campaigns of two weeks duration each, and a campaign of four weeks duration, covering a total period of 13 months. The objective was to investigate which meteorological conditions enhanced the probability of nucleation and to investigate any patterns in gaseous concentrations leading to the events8221;.

The corrected version is the following:

8220;This paper aims to analyse the frequency of and the atmospheric conditions favourable for nucleation events at coastal urban location in Brisbane, Australia, with a focus on the contribution of vehicle emissions. Monitoring was conducted during four campaigns of two weeks duration each, and a campaign of four weeks duration, covering a total period of 13 months. The objective was to investigate which meteorological conditions enhanced the probability of nucleation and to investigate any patterns in gaseous concentrations leading to the events to determine whether the local traffic was a major source of secondary particles in the study area8221;.

To maintain consistency, a new sentence was added between the first two sentences in the abstract. The original text was the following:

8220;The occurrence of and conditions favourable to nucleation were investigated at an industrial and commercial coastal location in Brisbane, Australia during five different campaigns covering a total period of 13 months. To identify potential nucleation events, the difference in number 5 concentration in the size range 148211;30 nm (N148722;30) between consecutive observations was calculated using first-order differencing8221;

8220;The occurrence of and conditions favourable to nucleation were investigated at an industrial and commercial coastal location in Brisbane, Australia during five different campaigns covering a total period of 13 months. The objective was to analyse the contribution of traffic-related emissions to the formation of secondary particles. To identify potential nucleation events, the difference in number concentration in the size range 14-30 nm (N14-30) between consecutive observations was calculated using first-order differencing8221;.

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Other corrections were made to the text, but these are detailed in our replies to the specific comments below

Specific Comments:

Comment 1:

The authors should carefully describe what they mean as a nucleation event. In most of the recent scientific literature the term 8220;nucleation event8221; has been used to refer to regional scale particle formation and growth that continues for hours (see e.g. Kulmala et al., 2004). I assume the 8220;nucleation events8221; referred to in this work are of a more local origin, if any? One example of particle formation events (there are others too) is the approach by Dal Maso et al. (Boreal Environ. Res. 10: 323-336, 2005). In most previous studies nucleation events are analysed mostly based on the full size distribution, taking also into account particle growth (rather than just bursts of small particles), whereas in this study the analysis seems to be done only based on particle numbers. Therefore, if the term 8220;nucleation event8221; is not carefully defined and compared to the literature use of the term, it is impossible (also for the authors themselves) to compare the results of this work to any previous studies on nucleation and particle growth

Response:

The sources quoted by the reviewer clearly make a distinction between 8220;nucleation8221; and 8220;particle growth8221;. However, the reviewer has rightfully pointed out that our definition of 8220;nucleation8221; needed to be clearer and consistent with the definitions provided in the literature. To address this observation, we made a change in the original text (page 2197, lines 3-5):

8220;Nucleation events, the formation of large numbers of particles at the lower end of the measurable size range, have been observed in different environments around the world8221;

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The corresponding correction to the above text is the following

8220;Nucleation events, that is, the appearance of a mode below 25-30 nm in the particle number size distribution, known as 8220;nucleation mode8221; (e.g. Dal Maso et al., 2007; Tunved et al., 2003), usually in very large numbers, have been observed around the world8221;.

To maintain consistency in our definition of nucleation, the first sentence in section 2.3

8220;The most important marker of a nucleation event is a significant increase in the concentration of nuclei mode particles in the time series8221;

has been changed. The new text now reads: 8220;Generally, secondary particles are in the size range < 30 nm (Morawska et al., 2008). Therefore, the most important marker of a nucleation event is a significant increase in the concentration of these particles in the time series. Needless to say, this increase is also reflected by the appearance of a mode in this size range8221;.

In addition, section 2.4 in the original text provides details of the techniques used to identify nucleation events, the atmospheric conditions favourable to these events and the classification of wind direction data in order to identify the origin or sources of these events.

Comment 2:

I have a serious concern about the choice of the measurement site and instrumentation in the aim of investigating nucleation events: It seems like a site that is very highly affected by different kinds of local emissions. This, on the other hand, makes it very difficult to assess whether secondary particle formation events take place or not 8211; particularly as the measurements start at 14 nm. I think it is very well possible that there are particles formed below 14 nm but they coagulate away before reaching detectable sizes. Also, it would be good to calculate the total particle surface area or condensational sinks and compare these to the concentrations in the smallest sizes to

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investigate a little bit the importance of coagulation losses at the site

Response:

This is a very valid comment and we addressed the issues conscientiously.

The reasons for selection of the site are added to the first two sentences in the second paragraph in section 2.1. The new text reads as follows:

8220;This site was selected because of the environmental diversity in the area. The sampling point was located between the road and railway line. The road and railway line follow a straight almost north-south path in the vicinity of the site8230;..8221;.

With regards to the concerns about the formation of particles below 14 nm, the reasons why no particles below this lower limit were measured were added to the text. The second paragraph of section 2.4 has been modified and now it reads:

8220;Due to limitations of the instrument, it was not possible to measure particles < 14 nm although the majority of these particles were expected to have formed below this limit. Therefore, it was assumed that the new particles were formed much earlier and grew principally by condensation mechanisms reaching a measureable size. Although losses due to coagulation were expected, most of these losses were likely to occur through coagulation with particles above 50 nm. Therefore, the appearance of a mode at < 30 nm indicated that a substantial number of new particles were formed allowing a significant fraction to grow to a detectable size. This means that nucleation events manifest themselves by an increase of particle number concentration in the 14-30 nm size range (N14-30). The first step was to divide the size distribution into 14-30, 30-50, 50-100, 100-300 and 300-800 nm and to calculate the total concentration of particles in each size class through the general formula:

(1)8221;

We did not consider necessary to calculate total particle surface or condensational sink, as they were beyond the scope the present study. Nevertheless, they could be

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directly deducted through visible increases in the number concentration of larger particles, which did not occur.

Comment 3:

Related to the two previous points, it would be nice if the authors would show plots of the evolution of the particle size distribution at the times when they have (or have not) defined a nucleation event to take place. Also, it would be interesting to know whether any particle growth is observed 8211; suggesting a regional scale phenomenon rather than just a burst of nanoparticles is observed. Differentiating between regional and local scale phenomena is important since their climatic importance is likely to be of very different magnitude.

Response:

Six plots were added, of the times, randomly selected, when nucleation events were identified (Figure 6 in the Appendix). A new paragraph was inserted to section 2.6 (last paragraph), which reads:

8220;Figure 6 provides samples of the evolution in particle size distribution during the nucleation events. In order to provide as much insight of the particle evolution during these events, only the hours when nucleation was observed are shown. The events showed similar patterns during each campaign. Therefore, only two samples per campaign, randomly selected, are shown. No nucleation was observed during the autumn campaigns.8221;

In addition, the paragraph in page 2206, line 26- page 2207 line 6 has been expanded and now reads:

8220;Figure 6 shows that the events were of very short duration, lasting a maximum of 4 hours. The typical 8220;banana8221; shapes observed in different studies (e.g. Boy Kulmala, 2002; Spracklen et al., 2006; Stanier et al., 2004) does not occur here. This means that the burst of N15-30 followed by growth was not observed and therefore

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these events are associated with air masses of local origin. Park et al. (2008) observed a similar phenomenon in the range 3-10 nm and hypothesized that this occurred because in the event nucleation and growth occurred above in the air and that subsequent vertical mixing caused the grown particles to reach the sampling point. They proposed this hypothesis after observing a gradual growth pattern only after particles larger than 10 nm had appeared. Although the lower size limit in our study was 14 nm, this hypothesis could be applied to this study provided that a growth pattern would be observed in the range above 30 nm. However, this did not occur. No increases in the number concentration of particles > 30 nm were observed after the events and the concentration of smaller particles dropped significantly after 2-4 hours. The analysis of the daily pattern of variation in size distribution and the concentration of size-fractionated particles indicates that the particle number is dominated by the range 50-100 nm, which is consistent with the size distribution observed for diesel exhaust emissions (Morawska et al., 1998). This is not surprising since the site is dominated by diesel traffic and sampling took place very close to the road. The dominance of this size range has the effect of increasing the available surface area. The presence of a high particle surface area prevents nucleation due to diffusion of small particles and condensed material to the surface of larger particles (e.g. Friedlander et al., 1991). This led us to hypothesise that air masses associated with nucleation events mixed quickly with emissions from the road. As a result, nucleation events were rare and of very short duration.8221;

Comment 4;

It is nice to have a long-term data set, but the authors should give detailed information on the exact times of the measurement periods. It should also be clearly defined what is meant by 8220;summer8221;, 8220;winter8221;, 8220;spring8221; or 8220;autumn8221;. Also, statistics on the goodness of the data (i.e. how many full days of data were collected and how large fraction of the data was reliable) should be given.

Response:

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The first paragraph of Section 2.2 has been expanded and now reads:

8220;Particle size distribution in the range 14-800 nm was measured with a TSI 3934 Scanning Mobility Particle Sizer (SMPS). The SMPS consists of a TSI 3071A electrostatic classifier (EC) and a TSI 3010 condensation particle counter (CPC). The EC classifies particles according to size while the CPC counts the number of particles in each size channel. An interfacing computer controls the process of measurement and stores the data supplied by the counter. Monitoring was conducted continuously at five-minute intervals during the following campaigns: autumn 2006 (12 May 8211; 4 June), winter 2006 (18 Aug-3 Sep), spring 2006 (31 Oct-15 Nov), summer 2007 (12-31 Jan) and autumn 2007 (23 May- 1 Jun)8221;.

The first paragraph of Section 2.5.1 was also expanded and now it reads:

8220;The diurnal patterns of mean variation in number size distribution in each of the different campaigns are shown in Figure 2. The data were highly reliable (> 95

Comment 5:

Related to the two previous points, it is very difficult for the reader to judge whether the mean size distributions given in Figure 2 are representative at all.

Response:

This is very related to the previous point. As shown in the previous response, more than 95

Comment 6: p. 2199, lines 18-20. The factor given in Eq. 1 is not really a correction but rather a normalization factor.

Response: In our previous publications this factor has been accepted as a correction factor. Nevertheless, its definition as a normalization factor is very valid and this definition has been adopted in the text.

Comment 7: p. 2199, lines 21-22. I supposed that the channels were logarithmically

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evenly spaced?

Response: To clarify this point, the paragraph after eq (2) has been expanded and now it reads;

8220;The SMPS data covered 64 channels per decade, evenly spaced in the logarithmic scale. Calculation of the log differences between consecutive size channels gave an average 61537;-value of 0.0156258221;.

Comment 8: p. 2200, lines 5-6. I supposed there should be a rather than a in Eq. 3 and the following line.

Response: is the symbol adopted in time series differencing whereas is used to calculate the rate of change over time. We were using time series differencing and therefore is the symbol that applies here.

Comment 9:

Why are there no signs of morning and evening rush hours in the traffic intensities? Is this specific to the site?

Response:

A final sentence has been added to the paragraph in page 2203, lines 3-9:

8220;The traffic pattern reflects the level of commercial activity in the area, which is high during the normal working hours8221;.

Technical Comments

Comment 10:

The figures are almost impossible to read because the size of the font is so small

Response:

The font size has been expanded (see Appendix 2). Comment 11: The map of the mea-

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surement site is confusing: again, the font size is too small, and the resolution of the image is too poor. Also, in the text the authors refer several times to a 8220;road8221; and 8220;railroad8221;. These should be clearly indicated in the map.

Response:

The map has been corrected (see Appendix 3). This is a colour map and was the best available for the area.

Comment 12:

Many of the references are not appropriate for the places where they appear. I suggest the authors carefully check all the references they cite.

Response:

The paper has been proofread and all the references were carefully re-checked for consistency with the citations. We couldn8217;t detect any errors. It is possible that the reviewer did detect the errors but because no further details were provided it was impossible to find those errors despite our best efforts.

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