Responses to referee#1's comments

Measurements:

Comment 1

There is practically no information on the experimental setup. What was the length of the sampling tubing, its material, the sampling flow rate? These are important, because PNC is very sensitive to diffusional losses in the ultra-fine range.

Response 1

Section 2.3 Measurement techniques. The following sentences have been inserted after sentence 1. "*Ambient air was drawn through a ~1m long conductive tubing connected to the EC. The ratio of the aerosol/sheath air flow for the EC was kept at 1/10 (0.6 to 6L min^{-1}), and the scan time was five minutes.*".

Comment 2

PNC at the QUT site was measured with an SMPS. The authors state that it consisted of an electrostatic classifier (TSI 3080), but do not specify whether a long or a short DMA was used. What was the sheath flow rate? How confident are the authors in the 4-110 nm size range they claim for the SMPS, when the CPC that they use (TSI 3781) has a nominal lower size limit of 6 nm?

Response 2

The type of DMA used in Electrostatic Classifier has been added.

Section 2.3, Measurement techniques, sentence 1, "UFP size…Condenstation Particle Counter (CPC) (TSI 3781)." has been revised to "*UFP size distribution in the range 4-110 nm was measured at the QUT monitoring site using a Scanning Mobility Particle Sizer (SMPS), which consisted of two parts, an Electrostatic Classifier (EC) (TSI 3080) equipped with a nano-Differential Mobility Analyser (nano-DMA) and a Condensation Particle Counter (CPC) (TSI 3781).*".

The sheath flow rate of Electrostatic Classifier has been added. Please refer to the response of comment #1, above.

The operation manual of CPC (TSI 3781) states that the D_{50} value is 6nm, which means that the detection efficiency of the particles with size of 6nm is 50%. According to the detection efficiency data of CPC (TSI 3781) provided by the manufacturer, that CPC can measure the smaller particles but with detection efficiency less than 50%. As a procedure of particle size distribution measurement, the particle number concentration of specific size is corrected by the detection efficiency factor (which is calculated by the SMPS software during the measurement). In the present study, the lower detectable size of the particles was down to 4 nm in the SMPS measurement. Therefore, we set the lower limit of the size distribution data to be 4 nm in our study.

Comment 3

The SMPS size range used in this study puts under question comparability of PNC measured with the SMPS and PNC measured with standalone CPCs used at the other two sites. Were there no particles larger than 110 nm at any of the three sites? These larger particles would be counted by the CPCs, but missed by the SMPS. Could this be part of the reason for the observed lack of correlation between the three sites?

Response 3

Although the SMPS measurements cover only the particle size from 4 to 110 nm, the

PNC obtained in this range is dominated in the study region by particles in this range (Morawska et al., 1998).
However, to remove the discrepancy of the measurement of PNCs by SMPS and CPC, a relative fraction of PNC to total PNC has been used in comparison and correlation analysis in this study, rather than an actual PNC. Therefore, the comparison between the SMPS and CPC measurements in this study are justifiable.

Clarification about the discrepancy of the two PNC measurement methods has been added.

Section 2.4, the following sentences have been added after sentence 4.
"*Since the PNCs measured at three sites were using SMPS and CPC, to remove the discrepancy of these measurement methods, a relative PN contribution to total PNC has been used in temporal and correlation analysis. Inter-comparison between the PNCs measured by SMPS and CPCs has been shown in* ***Figure-S1****; moderate correlations have been obtained (r^2 = 0.47- 0.81) with slopes of 0.55-0.65. This implies that the method of using PNCs measured by SMPS and CPCs for correlation analysis is justifiable and the ultrafine particles accounted for more than 50% of the PNC (by using CPC).*"

Comment 4
Were the SMPS and the two CPCs inter-compared side-by-side? Without such data there is very little value in any inter-correlation of PNC measured with these instruments.
Response 4
The two instruments are inter-comparable site by site and the information has been added about this in supplementary material.

Please refer to response to comment #3, above.

Data processing and analysis:
Comment 5
If I understand the 2nd sentence of section 2.4 correctly, SMPS channels with less than 1 cm^{-3} were discarded. This would be a wrong approach to treat SMPS data. Did you mean the integrated count (i.e. all channels combined)? How much did each of the quality control criteria contribute to the total 28% data removal?
Response 5
During the data processing process, we discard the data if the total PNC is less than 1 cm^{-3} (integrated count).

The contributions of data removal by each quality control criteria have been added.

Section 2.4. Sentence 3, "Approximately 28%...instrument malfunction." has been revised to "*Approximately 28% of the data removed from the database was based on the following criteria (the contribution of each quality control is shown in brackets): i) if the particle concentration has a zero value (~2%); ii) if the particle concentration is higher than 5 x 10^5 cm^{-3} (<1%); iii) and if data has been collected during instrument malfunction (~26%).*".

Comment 6

I find it hard to understand the rationale that the authors use for attributing PNC to different sources. For example, in the abstract they state that morning traffic exhaust emissions… contributed 5.5% and 5.15 (???) during the week respectively". This attribution seems to be based solely on the fraction that the morning rush hour concentrations represent of the total cumulative daily PNC. The origin of this is not explicitly discussed in the text, but becomes apparent upon examination of Fig. 2, 5, and 7. Unfortunately, this approach is simply naïve, because PNC is a combination of contributions from different sources, including the background. At the very least, the background contribution needs to be subtracted to estimate the traffic contribution. If the background has a diurnal pattern, as the authors seem to suggest (due to photochemical activity and new particle formation), this task becomes very problematic. A similar problem is found in the attribution of higher PNC observed in NE wind sector to industrial emissions (section 3.3). I do not understand how the authors can distinguish industrial emissions from nucleation events associated with that wind sector, which apparently happen very often (p. 32975, last sentence).

Response 6

We agree that the contribution of the total PNC by a specific source should be subtracted from the background concentrations. The discussion of morning vehicle emissions and photochemical production of particles in diurnal profile has been revised.

In abstract, line 7, "The results showed…contribution on weekends." has been revised to "*During the morning traffic peak period, the highest relative fraction of PNC reached about 5% at QUT and WOO on weekdays*.".

Section 3.1, the second paragraph has been re-written to "*In **Figure 2a,** it can be seen that morning PNC peaks were observed both at QUT and WOO. During that period, the measured relative fraction of PNC was found to be nearly 5% for both sites, however they were not found at ROC. This result suggests that the observed peaks are related to morning traffic activity on nearby roads. Around noon, PNC peaks were observed at all three locations, as well as the maximum solar radiation. The highest relative fraction of total PNC at noon is 7.6%, 6.0% and 8.9% for QUT, WOO and ROC locations, respectively. These peaks are likely to be the result of new particle formation caused by photochemical reactions (Cheung et al., 2011). It should be noted that the relative fraction of the total PNC is affected by background PNC, traffic emissions and photochemical particle production during the morning and noon periods. The maximum PNC observed at QUT and WOO is at 12:00, and at 13:00 for ROC. The time lag at ROC could be the result of the time the pollution plume requires to be transported from the upwind area (CBD area), to the downwind area (ROC). This is discussed in more detail in Section 3.5.*"

Comment 7

The explanation at the end of section 3.4 that the authors offer for the absence of nucleation event on 9 Sep 2009 at one of the sites (WOO) is less than convincing. The authors try to explain the absence of the event by a PM10 concentration that was higher by 30% than on the date when nucleation was observed at all three sites. First, it is doubtful that a 30% increase in PM10, which is a very poor parameter for condensational sink, would have such a dramatic effect to switch off the nucleation. Second, if the event was regional, then particle should be also forming upstream of

the site and should have been still arriving at the WOO site. Yet, apparently, it was not happening. The only explanation could be a rapid coagulation scavenging of the small particles by the larger ones. However, given very low total number concentrations (a few thousand per cm3), this seems highly improbable. Maybe the authors could provide an order of magnitude estimate of the time scales of coagulation losses?

Response 7

Since we do not have the particle size distribution data for WOO, it will be difficult to estimate the coagulation losses.

Section 3.4.1, paragraph 3, the following sentence has been amended to comment on the influence of coagulation scavenging. *"Beside the influence of condensation sink on the suppression of nucleation, the coagulation scavenging may have been another factor removing the freshly formed particles in this case."*

However, in our previous study (Jayaratne et al, 2011), is has been shown how an increase in PM_{10} concentration in the environment leads to a suppression of the ultrafine particle number concentration.

We have included the following sentence in Section 3.4.1, paragraph 3: "*A higher number of pre-existing particles in the atmosphere can act a strong sink for condensation nuclei, therefore suppressing the new particle formation (Kerminen et al., 2001). In our previous study (Jayaratne et al, 2011), it has been shown that an increase of PM_{10} concentration in the environment leads to a sharp decrease in the number of ultrafine particles".*

With the amended text, the paragraph at the end of Section 3.4.1 of the manuscript has been revised to: "*In terms of particle mass, the PM_{10} at WOO was 13.6 mg cm^{-3} on 17 July 2009, compared to 17.8 mg cm^{-3} on 9 September 2009 (an increase of 30.9%). On the other hand the PM_{10} at ROC was 7.0 mg cm^{-3} on 17 July 2009, compared to 6.8 mg cm^{-3} on 9 September 2009 (an decrease of 2.9%). A higher number of pre-existing particles in the atmosphere can act a strong sink for condensation nuclei, therefore suppressing new particle formation (Kerminen et al., 2001). In our previous study (Jayaratne et al, 2011), it has been shown that an increase of PM_{10} concentration in the environment leads to a sharp decrease in the number of ultrafine particles. Therefore, the relatively higher particle mass concentration of WOO on 9 September 2009, which is indicative of more pre-existing particles. This explains the suppression of the nucleation process at WOO on this day. Beside the influence of condensation sink on the suppression of nucleation, the coagulation scavenging may be another factor which removed the freshly formed particles in this case."*

The new reference to be added to the end of the manuscript is as follows:

CHARACTERISTICS OF AIRBORNE ULTRAFINE AND COARSE PARTICLES DURING THE AUSTRALIAN DUST STORM OF 23 SEPTEMBER 2009. E.R. Jayaratne, G.R. Johnson, P. McGarry, H.C. Cheung and L. Morawska. Atmos. Environ. 45, 3996-4001, 2011.

Minor comments:

Comment 8

p.32966, line 9: replace "5" with "%".

Response 8
p.32966, line 9: the number "5" has been replaced with "*%*" symbol.

Comment 9
p.32966, line 2: Remove "alteration"
Response 9
The word "alteration" is specific to describe the impact of aerosol on climate forcing, therefore it needs to be in this sentence.

Comment 10
p. 32966, line2: I doubt ultrafine particles contribute to visibility degradation they are in the Rayleigh scattering regime.
Response 10
In comment p.32966, line 2, the word "ultrafine" has been removed.

Comment 11
p.32967, line 19: There were several studies observing regional nucleation events in urban areas, such as Atlanta and Pittsburgh. Those studies were more comprehensive than the current one and need to be cited here.
Response 11
Previous studies on urban regional nucleation events of Atlanta and Pittsburgh have been citied.

Section 1, second paragraph, the last sentence, "For example…during the nucleation events." has been revised to "*For example, although regional nucleation has been observed in Helsinki, Finland (Hussein et al., 2008), Atlanta and Pittsburgh, United States of America (Stolzenburg et al., 2005, Stanier et al., 2004), spatial variations in PNC have also been studied in urban areas in Australia (Mejia et al., 2008) and in the United States (Hudda et al., 2010). These studies have not examined the impact of regional pollution on PNCs or the influence upwind urban pollution has on PNC downwind during the nucleation events.*"

Comment 12
p.32969, line 5: How "heavy" was the traffic flow? Provide traffic count.
Response 12
A traffic count has been added to p.32969, line 5, "a road with heavy…Brisbane to the CBD." has been revised to "*a road with a heavy traffic flow volume of over 40,000, connecting the Southern Brisbane suburbs to the CBD.*"

Comment 13
p.32970, line 6-7: The TEOM measurements have been already mentioned in the previous paragraph.
Response 13
In p.32970, line 6-7, the sentence "a tapered element oscillating microbalance (TEOM)." has been removed.

Responses to referee#2's comments

Comment 1
Page 3, Line 14. Although the authors have acknowledged in the introduction that size distributions of PNC have been less investigated, and the present study did employ a SMPS for size distribution measurement at least at one site, such information was not presented at all in the figures and discussions. Rather, only PNC was calculated based on the sum of all size channels from SMPS. The temporal variation of the particle size distribution could carry important information of UFP formation and evolution. It is highly recommended to include the data and discussion in the manuscript.
Response 1
In this manuscript, we aim to focus on the regional variation of PNC at three locations in Brisbane, Australia during nucleation events of the year 2009. Discussions on particle size distribution and new particle formation for this measurement period have been included in another article titled "*Observation of new particle formation in subtropical urban environment* (Cheung et al., 2011)", published in Atmospheric Chemistry and Physics, 11, 3823-3833. This reference has been included in the manuscript, therefore there is no need to include this discussion in this article as well.

Comment 2
Page 3 Line 2. "thay are" is a typo and it should be "they are".
Response 2
On page 3, line 2 the word 'thay' in "thay are" has been revised to "*they are*".

Comment 3
Page 5, Line 16. The measurement techniques part is not clear. CPC 3781 measures particle with size >6 nm and the authors stated the size distribution measurement with TSI 3080 + CPC 3781 tandem was in the range of 4-110 nm. Such discrepancy of information needs clarification. Also the type of DMA used isn't clear and from the presented information, it seems to be 3080N, a nano-DMA. It needs confirmation.
Response 3
In depth information about particle size distribution measurements have been added in the Section 2.3 (measurement techniques) of the manuscript.
In section 2.3, paragraph 1, sentence 1 the sentence "UFP size distribution…for the QUT site." has been revised to "*UFP size distribution in the range of 4-110 nm was measured at the QUT monitoring site using a Scanning Mobility Particle Sizer (SMPS), which consists of two parts: an Electrostatic Classifier (EC) (TSI 3080) equipped with a nano-Differential Mobility Analyser (nano-DMA) and a Condensation Particle Counter (CPC) (TSI 3781).*"

In regards to the discrepancy about PNC measurements by SMPS and CPC, please refer to response to referee #1, comment #3, above.

Comment 4
The second paragraph of the section didn't mention where the real time measurements were located. It seems to be WOO and ROC with DERM stations. Also the information TEOM was repetition from the first paragraph.
Response 4
Locations of real time measurement have been added in the manuscript in section 2.3, paragraph 2, sentence 1, "Gaseous pollutants…for O3)." has been revised to

"*Gaseous pollutants, such as carbon monoxide (CO) and nitrogen oxide (NO_x), were measured at WOO; and ozone (O_3) and CO were measured at ROC using real-time gaseous analysers (Ecotech ML9830 for CO; Ecotech ML9841/ API 200A for NO_x; Ecotech ML9812 for O_3).*"

In section 2.3, paragraph 2, sentence 2, the sentance "Meteorological parameters…" has been revised to "*Meteorological parameters, including wind direction/speed, temperature, relative humidity and solar radiation, have also been measured.*"

Comment 5
Page 6, Line 12. It is stated that approximately 28% of the data were removed based on the given criteria. The ratio of outlier seems quite high, and it is recommended to give possible reasons for this for readers' reference.
Response 5
Please refer to response to referee #1, comment #5, above.

Comment 6
Page 7, Result and discussion. It is a big concern for the direct comparison of SMPS derived PNC (4-110nm as stated) with CPC 3781 measurement (>6 nm) form different locations. First SMPS tandem uses a 3781 CPC with size cut >6nm so how effective was the distribution measurement for particles size <6nm; Second, for the upper size ranges > 100nm. It is assumed in the manuscript that their contribution is negligible and the PNC from both set up were compared without justifications. If the authors have collected side-by-side data for both CPC 3781 and SMPS (DMA 3080N + CPC 3781) during the study, it is recommended to present examples and analysis to clarify the concerns.
Response 6
In regards to the discrepancy about PNC measurement by SMPS and CPC, and side-by-side SMPS and CPC comparison, please refer to response to referee #1, comment #3, above.

Comment 7
Page 7, line 9. For the presentation of grant average of diurnal profiles of the PNC from three sites, it is not clear if the averages were based on the overlapping periods of the measurement or the individual measurement. The roadside site missed almost half of the sampling time compared with the other two. It is recommended to make clarifications and justify the data presentation.
Response 7
The diurnal profiles of PNC have been obtained from the data collected from each site, but not during the overlapping periods between the three locations.
Clarification about the data collection has been added in section 2 (Methods and Techniques).

In, Section 3.1, the following sentence has been inserted after sentence 1: "*It should be noted that the measurement periods at each site did not overlap."*

In section 2.4. paragraph 1, the following sentence has been amended:
"*It should be noted that the PNC data for WOO is missing for the months from January to April due to instrument malfunction.*".

Comment 8
Page 7, line 18, it is not clear how 5.5% and 5.1% were calculated to evaluate the contributions from morning PNC peaks. If it is based on the ratio of peaks values over the sum of PNC from all day measurements, it may be questionable. The authors need to clarify the method of calculation. Same comments go to the relative contribution calculations for noon peaks.
Response 8
Please refer to response to referee #1, comment #6, above.

Comment 9
Page 8 line 2, in the discussion of the diurnal trend of PNC, it is suggested to describe the temperature profiles during the sampling period since the stated pattern seems to be the impact of different heights of mixing layer that has been observed in most of the urban environments. However, only temperatures of two days of case studies were presented with little variation. How about other days?
Response 9
Similar observations with high r^2 (QUT-ROC) and relatively low r^2 (QUT-WOO), r^2 (WOO-ROC) during the case study on 9 Sep 2009 are found on the days 30 May, 8 June, 8 August and 16 October. On these days, temperature differences during the nucleation events between WOO and ROC are found to be 1.4 ºC, 1.7 ºC, 1.5 ºC and 0.6 ºC, respectively, showing that the heights of mixing layer (ie temperature) at these locations are similar. Based on our data, the suppression of new particle formation at WOO occurred at several days (30 May, 8 June, 8 August, 16 October and 9 Sep 2009) and in these days the temperature (ie. the mixing height) at WOO and ROC are almost the same, and hence temperature is not a major factor to the observed difference in particle formation.

In section 3.4.1, the following sentence has been amended:
"*The temperature differences during those events at WOO and ROC were small, implying that the impacts of height of mixing layer on both locations were similar. This observation (with high r^2 (QUT-ROC) and relatively low r^2 (QUT-WOO), r^2 (WOO-ROC)) was also found on 30 May, 8 Jun, 8 Aug and 16 October 2009.*".

Comment 10
Page 10, line 4. For the discussion of the correlations for nucleation events, these are interesting findings, but it is also possible that the high correlation coefficients were actually driven by the much higher values concentrations from all sites due to the burst PNC. What are the p values of the correlations? It is recommended to show examples of actual scatter plots of nucleation and non-nucleation events in the SI to strengthen the point.
Response 10
The correlation were coefficients of PNCs between the different locations presented in Table 2, selected for the days with nucleation growth events only (except the case on the 15 March, during which the nucleation growth event was observed in the morning, and burst events in the afternoon) which do not include any nucleation burst events. The *p* values of the correlations calculated are <0.05. The examples of actual scatter plots (**Figure S2**) of nucleation (17 July 2009) and non-nucleation (8 March 2009) have been added in the supplementary material.

In section 3.4, first paragraph, the sentence "For example…respectively." has been revised to "*For example, the r^2 values for QUT-ROC, QUT-WOO and WOO-ROC were 0.95, 0.71 and 0.75 on 17 July 2009 respectively (with $p < 0.05$).*".

In section 3.4, first paragraph, the following sentence has been amended: "*To better illustrate the correlation of PNCs from different locations (urban and downwind semi-urban areas) during nucleation and non-nucleation events, an example of the PNC scatterplot obtained during the event and non-event days at QUT and ROC is shown in* ***Figure S2***."

Other revisions

On p.32966, line 5-8, the sentence "Due to their small…" has been revised to "*Due to their small size (< 0.1 μm), ultrafine particles (UFPs) only contribute a very small amount to the total mass of atmospheric particles, however they are most abundant by number (~70-90%) and potentially have a greater impact on human health than the larger particles (< 2.5 μm) (Morawska et al., 2008).*"

In section 2.3, measurement techniques, sentence 2, the sentence "The size distribution…at each site." has been revised to "*The size distribution data is then used to calculate PNC for the QUT site. At the WOO and ROC stations, PNC is continuously measured by a water-based CPC (TSI 3781) with a size-cut inlet of 1 nm, while particle mass concentrations of $PM_{2.5}$ and PM_{10} are measured by a Tapered Element Oscillating Microbalance (TEOM) in 30-minute intervals at each site.*".