Responses to referee#2's comments

Comment #1

Page 8993: Based on nighttime measurements, the authors have derived a primary emission ratio of 60 cm-3 per ppbv of NOx emissions. I suppose a corresponding relation have been reported in some other investigations as well. How does this number compared with values found by others?

Response #1

Up to our knowledge, there have some similar results reported for the total particle number concentration (N_{total}) compared to the NO_x. For example, 225-661 cm^{-3}/ ppbv (N_{total}/NO_x) obtained in urban areas of United Kingdom (Harrison et al. 2005), and about 160-345 cm^{-3}/ μg m^{-3} (~ 302-651 cm^{-3}/ppb) for urban Stockholm (Gidhagen et al. 2005). However, the relationship between N_{nuc} and NO_x has never been reported. One plausible reason is that the particle size distribution of vehicle exhaust emission was usually dominated in the range of 20-130 nm (Morawska et al. 2008) and, therefore, the data analysis focus on the relationship between N_{nuc} and NO_x was rare. Thus, no comparison between this result and other studies was discussed in the manuscript.

Morawska, L., Ristovski, Z., Jayaratne E.R., Keogh, D.U. and Ling, X.: Ambient nano and ultrafine particles from motor vehicle emissions: Characteristics, ambient processing and implications on human exposure. Atmospheric Environment, 42, 8113-8138, 2008.

Harrison, R.M. and Jones, A.M.: Multsite Study of Particle Number Concentrations in Urban Air. Environmental Science and Technology, 39, 6063-6070, 2005.

Gidhagen, L., Johansson, C., Langner, J. and Foltescu, V.L.: Urban scale modeling of particle number concentration in Stockholm. Atmospheric Environment, 39, 1711-1725, 2005.

Comment #2

Page 8994 and figure 5: How was Aitken mode particle number concentration changing diurnally? The discussion and fig 5 might benefit from including Aitken mode particles as they connect nucleation and growth. Lines 9 and 10: technically, total particles cannot be divided into nucleation and accumulation mode particles, as there is always Aitken mode present as well.

Response #2

-Figure 5 has been revised which include the Aitken mode particle number

concentration.

-Page 8994, section 3.4, paragraph 1, line 5. From the sentence "Averaged diurnal variations…" until the end of this paragraph have been revised to:

"*Averaged diurnal variations of N_{total} and UVB for this study are depicted in **Figure 5**. Two peaks were observed for N_{total} at 11:00 LT and 18:00 LT. The first one was most likely contributed by NPF which was associated with daily maxima of UVB, whereas the second peak showed the influence of traffic emission in late afternoon. We then separate the N_{total} into three size bins as described in section 2.3, and the diurnal variations in the number concentrations for nucleation and Aitken mode particles (N_{nuc} and N_{Aitken}) , are also illustrated in **Figure 5**, respectively. A mode of N_{nuc} was observed near 10:00 LT and the peak of N_{Aitken} was observed one hour later. This suggested that nucleation mode particles were formed in the morning where particles have subsequently grown into larger sizes. (Cheung et al., 2011).*".

-Caption for figure 5 has been revised.
"***Figure 5.** Diurnal variation of particle number concentrations of total (N_{total}, red solid line), Aitken (N_{Aitken}, blue dash line), and nucleation (N_{nuc}, green dash line) modes in lower panel, also UVB index in upper panel.*".

Comment #3
Page 8995, line 1: What do the authors mean by stating that NPF influencing significantly on local air quality? The particle number is usually not used as a measure of air quality, and NPF did definitely not have any influence of particulate matter concentrations.
Response #3
We agree that the mass fraction of nucleation mode particles in ambient PM is very small and thereby have very limited influences on the mass concentration of PM. However, it is still very likely that the increases in the number concentration of aerosols due to NPF could cause negative impacts to visibility and public health. To avoid confusion, we have made the following revision:

-Page 8994, line 28. The sentence "These results suggested…" has been revised to "*These results suggested that the burst of nucleation mode particles induced by the NPF had caused a significant impact on the particle number concentration during this campaign period.*".

Comment #4

Page 8996: The interpretation of figure 9 is weak. Without that one point at the upper part of the plot, a linear fit to the data would be as good as exponential one. The authors have excluded one point from the bottom of the same plot without a good reason. In my opinion, the only thing that can be interpreted from the figure is that GR increase with increasing J(O1D). Claiming that the increase is exponential is not convincing.

Response #4

We agree that the interpretation of figure 9 is rather weak. We excluded the case of 11 July for relatively high wind speed; however, we did not find a reason to exclude the case of highest GR. Indeed, if we treat the extreme case as an outlier, the correlation can be depicted using either linear or exponential fit. We selected using exponential fit because we want to demonstrate that the relationship between GR and ozone photolysis could be nonlinear. The thought is that production of OH radicals can contribute to GR not only for the production of condensable vapors that condense onto particles, but also via formation of new tiny particles that could coagulate with pre-existing particles and enhance the apparent growth rate. Thus, we hope we can keep the figure and revise the description as the followings:

*"**Figure 9** illustrates the correlation between GR and the product of $J(O^{1D})$ and mixing ratio of ozone, which gives the production rate of OH radicals from ozone photolysis. It was revealed that the value of GR increased with photolysis of ozone and in turn the production of OH radicals. The relationship between GR and ozone photolysis is depicted exponentially and suggests that the GR could be changing nonlinearly with OH production. One plausible explanation of the nonlinear relationship is that production of OH radicals can contribute to GR not only for the production of condensable vapors that could condense onto particles, but also via formation of new tiny particles that could coagulate with pre-existing particles and enhance the apparent growth rate. Thus the significant correlation between GR and OH production warrants that the growth of newly formed particles was driven by production of condensable vapors, either organics or H_2SO_4, from photochemical reactions. However, as the data from this study characterized the relationship between GR and ozone photolysis, further studies to link the photochemical dynamics and microphysical behavior of aerosols are needed to validate the hypothesis of nonlinear growth. The outlier shown in **Figure 9** is for the event on 11 July. The causes of that discrepancy are yet unclear; nevertheless, preliminary investigation suggested that the growth of particle could have been inhibited by stronger winds during that morning (shown in **Figure S4**)."*

Section 3.5: There are a few minor issues related to formation and growth in this section that could be improved.

Comment #5

First, I recommend that the authors use J10 instead of J10-25, i.e. the formation rate of 10 nm particles, to make the notation consistent with most of the literature. The upper limit is not that relevant here.

Response #5

-The notation of $J_{10\text{-}25}$ has been revised to J_{10} in the manuscript.

Comment #6

Second, I do not agree that the observed formation rates of 10 nm particle are at the lower limit of values reported in the literature. It is true that J10 is comparable or larger than observed here in many locations, but in rural or more remote location values of J10 are almost always lower than reported here. Besides the Kulmala et al. 2004 paper, the authors could include more recent material on J and GR (see e.g. Manninen et al. 2010, Atmos Chem Phys, 10, p 7907-7927, and reference therein).

Response #6

Since the particle number concentration was re-calculated therefore the corresponding discussion on the particle formation and growth rates was revised.

-Abstract has been revised.

"Averages (± 1σ) of the diameter growth rate, GR, and formation rate of nucleation mode particles, J_{10}, were 11.1±10.4 nm h^{-1} and 6.2±2.9 cm^{-3} s^{-1}, respectively."

-Section 3.5, paragraph 2. From "Averaged formation rate…in urban Beijing, China (Yue et al., 2010)." has been revised.

"*Averaged formation rate of nucleation mode particles (J_{10}) for each NPF event was calculated for the particles size ranged from ~10 to 25 nm according to the method of Dal Maso et al. (2005). Formation rate is defined as sum of the apparent formation rate (dN_{nuc}/dt) and the coagulation loss rate during the NPF event. It should be noted that the reported apparent particle formation rate (J_{10}) is expected to be smaller than the actual nucleation rate (or the formation rate of 3 nm particle, J_3), since some fraction of formed nuclei is always scavenged by coagulation into larger pre-existing particles before they grow larger by condensation (Lehtinen et al., 2007). The mean J_{10} for the new particle events was found to be 6.2±2.9 cm^{-3} s^{-1}. The J_{10} observed in this study ranged from 1.4 to 12.0 cm^{-3} s^{-1}, which is comparable to recent observation for NPF in urban areas such as Budapest, Hungary (J_6: 4.2 cm^{-3} s^{-1} with a range of*

1.65 – 12.5 cm^{-3} s^{-1}, Salma et al., 2011), Beijing, China (J_3: 2 – 13 cm^{-3} s^{-1}, Yue et al., 2010), Marseille, France (J_3: 3 – 5.3 cm^{-3} s^{-1}, Petäjä et al., 2007) and Athens, Greece (J_3: 1.3 – 6.5 cm^{-3} s^{-1}, Petäjä et al., 2007).”.

-Section 4, last paragraph, last sentence “In this context…” has been deleted.

-The following references have been added into the reference list.

Petäjä, T., Kerminen, V.-M., Dal Maso, M., Junninen, H., Koponen, I.K., Hussein, T., Aalto, P.P., Andronopoulos, S., Robin, D., Hämeri, K., Bartzis, J.G. and Kulmala, M.: Sub-micron atmospheric aerosols in the surroundings of Marseille and Athens: physical characterization and new particle formation. Atmospheric Chemistry and Physics, 7, 2705-2720, 2007.

Salma, I., Borsós, T., Weidinger, T., Aalto, P., Hussein, T., Dal Maso, M. and Kulmala, M.: Production, growth and properties of ultrafine atmospheric aerosol particles in an urban environment. Atmospheric Chemistry and Physics, 11, 1339-1353, 2011.

Comment #7

Third, the authors should state explicitly that J10 is always smaller than the actual particle formation rate (nucleation rate), or the formation rate of 3 nm particle reported in some studies, because some fraction of formed nuclei is always scavenged by coagulation into larger pre-existing particles before they grow larger by themselves by condensation (see Lehtinen et al. 2007, J. Aerosol Sci. 38, p 988-994).

Response #7

-Discussion about the difference between J_{10} and J_3 (reported in other studies) has been added into the manuscript.

-Following sentences have been inserted after section 3.5, paragraph 2, sentence 2.

“*It should be noted that the reported apparent particle formation rate (J_{10}) is expected to be smaller than the actual nucleation rate (or the formation rate of 3 nm particle, J_3), since some fraction of formed nuclei is always scavenged by coagulation into larger pre-existing particles before they grow larger by condensation (Lehtinen et al., 2007).”*.

-Following reference has been added into reference list.

“Lehtinen, K.E.J., Dal Maso, M., Kulmala, M. and Kerminen, V.-M.: Estimating

nucleation rates from apparent particle formation rates and vice versa: Revised formulation of the Kerminen-Kulmala equation. Journal of Aerosol Science, 38, 988-994, 2007.".

Minor/ technical issues

Comment #8

Different units should be separated with a space (e.g. ug m-3 and m s-1 on page 8992).

Response #8

-Different units have been separated with a space in the manuscript.

Comment #9

Page 8989: please spell out a.g.l and Lpm.

Response #9

-The abbreviation of a.g.l and Lpm were requested by ACP format since those terms were commonly used. Therefore we would like to keep it in manuscript.

Comment #10

Page 8992, line 15: should read associated with…

Response #10

-Page 8992, line 15. "…associated to…" has been revised to "…*associated with.*…".

Comment #11

Page 8994, line 27: one fold sound strange in this context. Do the authors mean several fold or 10 fold?

Response #11

-Page 8994, line 27. "Also the 95th…" has been revised to "*Also the 95th percentile of N_{nuc} (24.3 x 10^{3} cm^{-3}) during the NPF event reached twice of that measured on non-event days.*".

Responses to referee#3's comments

General comments

Comment #1

The topic of this paper is of interest but some parts of the data analysis needs to be revised. This paper can be published in ACP but after the major revision considering the comments below.

Response #1

-Thank you for your comments on this manuscript. The manuscript has been revised according to the comments.

Specific comments

Methodology:

"Observation site and instrumentation" section:

Comment #2

Was the loss due to diffusion inside the SMPS system corrected? If yes, which methods were used? Did you find the diffusion loss of the particular system you used yourself or you applied the correction using the data available in literature? If no, why the loss due to diffusion was not corrected as your result can be influenced by diffusion loss particularly when you are considering particles in nucleation mode.

Response #2

-Diffusion loss inside the SMPS was not corrected in previous version of manuscript, and the correction has been applied for the current manuscript by using the internal diffusion loss scheme of TSI AIM software. Particle number concentrations have been re-calculated and corresponding results (including relevant tables and figures) in manuscript have been revised.

-Section 2.2, paragraph 2, last sentence "Multiple charge…" has been revised to "*Multiple charge and diffusion loss corrections were applied to the particle size distribution measurements using the internal algorithm from the Aerosol Instrument Manager Software.*".

Comment #3

It is stated that the PSL test were conducted for the size accuracy test of the SMPS. Which PSL diameter(s) were used and why? How much error from the PSL nominal diameter was acceptable?

Response #3

-Two PSL particles were used for SMPS testing, which have nominal diameters of 97

± 3.7 nm (Part#: 3100A, lot#: 35431, Thermo Scientific Inc.) and 350 ± 6.0 nm (Part#: 3350A, lot#: 35910, Thermo Scientific Inc.). The testing results were 97.5 ± 0.3 nm and 341.9 ± 5.2 nm, respectively. The difference between the measured particle sizes and nominal diameters for PSLs were 0.5 % and 2.3 %. The less than 3% difference for the particle sizing accuracy for SMPS is reasonably.

-Section 2.2, paragraph 2, sentence 6. "Besides, the accuracy…" has been revised into "*Besides, the accuracy of the particle sizing of EC was checked using polystyrene latex spheres (PSL) before the campaign. The nominal diameters of the PSL were 97 ± 3.7 nm (Part#: 3100A, lot#: 35431, Thermo Scientific Inc.) and 350 ± 6.0 nm (Part#: 3500A, lot#: 35910, Thermo Scientific Inc.). The measured sizes of the PSL by the SMPS were found to be 97.5 ± 0.3 nm and 341.9 ± 5.2 nm, respectively. Less than 3% differences between the nominal and measured diameters were obtained.*".

Comment #4

How did you identify groups A and B in your data? Which method did you use in order to find the number of groups and divide the data into those groups? It seems that it was done only visually and no quantitative methods were applied to find the groups in data. How reliable this grouping can be if this is the case?

Response #4

-Firstly, the grouping was based on visual observation. Then further data analysis by separation of the data into daytime/nighttime was conducted (by assumption of vehicle exhaust emission dominated during the nighttime of urban environment). Figure 3 has been revised that nighttime data was showed in red color dot to make two groups easier to identify. Also corresponding discussion has been revised.

-Section 3.3 has been revised into

"*Relationship between PNC and other parameters were further assessed in this section. Pearson correlation coefficients, r, were calculated between PNC and particle mass concentrations and primary gaseous pollutants (i.e. NO_x and SO_2). The scatter plots between PNC and $PM_{2.5}$ are depicted in* ***Figure 3 (a-c)****. In general, PNC does not show a strong correlation against mass concentrations. For example, the r value between $PM_{2.5}$ and N_{nuc} was found to be 0.34 ($p < 0.05$). However, an obvious relationship between PNC and $PM_{2.5}$ was found when only the nighttime data was considered (highlighted in red color).*

To better examine the relationship between N_{nuc} and vehicle exhaust emission, N_{nuc} and NO_x were plotted in scatter plots for the daytime and nighttime, respectively, where the mixing ratio of NO_x was used as an indicator of local vehicle emissions.

*For the nighttime data (see **Figure 4a**), a significant linear correlation between N_{nuc} and NO_x was obtained ($r = 0.87$, $p < 0.05$), suggesting that vehicle exhaust emission was the predominant source of nucleation mode particles. The slope of 157.5 cm^{-3} N_{nuc} per ppbv of NO_x characterizes the emissions from vehicles which contributed to the background level of nucleation mode particles in an urban area. Note that this emission ratio was applicable only for the study area, since it would depend on the local emission properties (i.e. types of fuel and vehicles) and meteorological conditions. **Figure 4b** shows the daytime data of N_{nuc} against NO_x with a line fit obtained from **Figure 4a** to represent the "urban background" N_{nuc} contributed by vehicles. Note that, at times, the daytime N_{nuc} was enhanced up to 10 times the level estimated by the N_{nuc}/NO_x emission ratio. The results suggest that, in the budget of N_{nuc} of urban Taipei, the primary (NO_x relevant) sources contributed throughout a day, whereas the other (NO_x independent) sources dominated during daytimes, in particular the episodes of new particle burst. Previous studies of NPF events in urban areas showed that the burst of nucleation mode particles was associated strongly with photochemical production of sulfuric acid (Woo et al., 2001; Stanier et al., 2004; Cheung et al., 2012). Similar finding has been reported in other polluted urban area of central Taiwan (Young et al., 2012). The discussion on the NPF will be provided in the following sections."*

Comment #5

It is stated that "Vehicle exhaust emission was suggested to be a major source contributing the group "A" pollution…" How did you come into this conclusion?

Response #5

-NO_x was used as an indicator of vehicle exhaust emission for nighttime data which shows a good linear relationship with N_{nuc}. See response for comment#4 for further detail discussion.

Comment #6

95% confidence interval needs to be plotted in Fig. 5.

Response #6

-95% confidence interval has been added for the particle number concentration.

Comment #7

Second peak in PNC were attributed to the afternoon rush hour. Why no peak was observed in the morning due to morning rush hour?

Response #7

-Figure 5 has been revised to include N_{total}, N_{Aitken} and N_{nuc}. The morning peak of PNC is existed for N_{Aitken} but weak compared to the afternoon peak.

Comment #8

UVB*SO2 was used as proxy to the H2SO4, however, this term does not consider the condensation sink (CS). CS needs to be calculated and UVB*SO2/CS should be used as a proxy for sulphuric acid[1].

Response #8

-The purpose of the proxy value used here is an indicator of the influence of H_2SO_4, but not actual approximation of N_{nuc} induced by H_2SO_4. If accurate approximation need to been calculated, the concentration of [OH] is needed, rather than the UVB.

On the other hand, I agreed that the effect of condensation sink should be accounted for new particle formation process. Therefore, the term "UVB*SO_2" has been replaced by "UVB*SO_2/CS" in figure 7, and corresponding discussion has been revised in manuscript.

-Section 3.4, paragraph 3 has been revised.

"During the NPF events, the variation of N_{nuc} was found to be coincident with an index of photochemical production of ambient H_2SO_4, which was defined by the product of UVB and SO_2 divided by the particle condensation sink (UVBSO_2/CS).* ***Figure 7*** *shows the time series of N_{nuc} and UVB*SO_2/CS (upper panel) and the particle size distributions from 8-14 July during which consecutive nucleation events were observed. The N_{nuc} and UVB*SO_2/CS was fairly correlated in log-normal relationship with r^2 ranging from 0.51 – 0.63 (see* ***Figure S1*** *in supplementary for the scatterplot between N_{nuc} and UVB*SO_2/CS). This implies that the increases of the N_{nuc} during the NPF events were also contributed by other sources, for example the local vehicle emission which was mentioned in Section 3.3. Nevertheless, the result suggested that H_2SO_4 was major contributor to the particle formation process in our study region."*.

-Caption for figure 7 has been revised.

"***Figure 7.*** *Time series of particle size distribution (lower panel); and number concentration of nucleation mode (N_{nuc}) and UVB*SO_2/CS (upper panel) for 8 – 14 July 2012.*".

Comment #9

It is stated that"…the variations of Nnucl and UVB*SO_2 were qualitatively agreed". Is it a right approach to correlate two sets of quantitative data qualitatively? Correlation should be calculated quantitatively using the proper statistical methods.

Response #9

-According to Comment #8, "UVB*SO_2" was replaced by "UVB*SO_2/CS" and corresponding discussion has been revised (see response for comment# 8). Correlation analysis between N_{nuc} and UVB*SO_2/CS has been conducted. Moderate log-normal relationship was found with r^2 ranging from 0.51 - 0.63 (see Figure S1 in supplementary). This result indicated that these two parameters have certain positive correlation.

Comment #10

It is also stated that"…results suggest the H2SO4 was playing an important role for the particle formation process…"How did you come to this conclusion? H2SO4 may be involved in the particle growth but may or maynot play a role in the formation based on your results.

Response #10

-Data result shows that a moderate log-normal correlation between N_{nuc} and H_2SO_4 proxy (i.e. UVB*SO_2/CS) has been found (see response #8). This indicated that the H_2SO_4 contributed in the particle formation process.

Other revisions.

1) Some grammar mistakes have been corrected in the manuscript

2) References in manuscript and reference list have been updated.

2) Section 2.2, paragraph 3. Instrument information of CO has been removed.