Referee #1

General comments:

In this study the seasonal variation of size distribution, mass, and chemical composition of ultrafine and submicron particles is investigated at an urban site in East Asia. Authors found seasonal differences in the particle size distributions: in summer Aitken and accumulation mode particle concentrations were higher than in other seasons, while the concentration of nucleation mode particles was lower. In addition, the mass concentration of ultrafine particles was observed to be highest in summer, and they were composed mostly of organic carbon. Sub-micron particles had the highest mass concentration in spring, and their major constituent was sulfate. Furthermore, new particle formation events were observed at the site during spring and summer.

I believe that this study is scientifically relevant and can be published in ACP after revisions. Authors should present some results on the diurnal variation of particle size distribution in different seasons. In addition, the structure of the "Results and discussion" section should be changed so that the paragraph on NPF events is not in the end of the section, and Table S3 should be moved to the main text. Furthermore, authors should show how high sulfuric acid proxy and low PM10 favor particle formation with some additional figures. The language of the manuscript should also be corrected (for example the tense of verbs should be checked). Moreover, many of the figures are difficult to read because of too small size/or low quality. More specific comments are presented below.

Response to General comments

Thank you for the comments and suggestions by the referee. The comments on the structure of manuscript, and the favor condition of NPF events have been revised. Figures in the manuscript have been revised to make it clearer and easier to read. The use of language of the manuscript has been modified. The responses for specific and technical comments have been listed below:

Specific comments

Comment #1

The title of the manuscript should include the information on the measurement site.

Response #1

The title has been revised to "*Seasonal variations of ultrafine and sub-micron aerosols in Taipei, Taiwan: implications for particle formation processes in urban areas*"

Comment #2

Page 21805, line 13: Newer references should be added here.

Response #2

New references have been added, which are as follows:

Kulmala, M., Petäjä, T., Nieminen, T., Sipilä, M., Manninen, H.E., Lehtipalo, K., Dal Maso, M., Aalto, P.P., Junninen, H., Paasonen, P., Riipinen, I., Lehtinen, K.E., Laaksonen, A. and Kerminen, V.-M. (2012). Measurement of the nucleation of atmospheric aerosol particles. Nature Protocols, 7, 1651-1667.

Comment #3

Page 21805, line 16: Here it would be good refer to some of the review papers on particle formation.

Response #3

The cited references in this part have been replaced by two review papers below:

Kulmala, M., Vehkamäki, H., Petäjä, T., Dal Maso, M., Lauri, A., Kerminen, V.-M., Birmili, W. and McMurry, P.H. (2004). Formation and growth rates of ultrafine atmospheric particles: a review of observations. Journal of Aerosol Science, 35, 143-176, 2004.

Holmes, N.S. (2007). A review of particle formation events and growth in the atmosphere in the various environments and discussion of mechanistic implications. Atmospheric Environment, 41, 2183-2201.

Comment #4

Page 21806, line 6: The meaning of the sentence starting with “In a subtropical urban area…” is not clear for me.

Response #4

Page 21806, line 6: The sentence of “In a subtropical…” has been revised to “*In Taipei, Taiwan, a subtropical urban area, Cheung et al. (2013) observed that there were a ten-fold increase in nucleation mode particle number concentrations ($N_{9\text{-}25}$, with size 9 < d < 25nm) during new particle formation events compared to that contributed by the vehicle emission.*”

Comment #5

Page 21807, line 20: Instead of “electrostatic mobility” a term “electric mobility” is more commonly used.

Response #5

Page 21807, line 20: The term “electrostatic mobility” has been revised to “*electric mobility*”.

Comment #6

Page 21809, line 24: More details about trajectory calculations should be added. It is not clear if trajectories were calculated for each hour of the measurement period or less frequently. The arrival height of the trajectories should also be mentioned.

Response #6

The detail information about the trajectory calculation has been added.

Page 21809, line 20: The following sentence has been inserted after the first sentence.

"*72-h back trajectories were calculated twice per day at 00:00LT and 12:00LT with height setting of 200 m above ground level.*".

Comment #7

Page 21809, line 25: This paragraph should be moved to "Results and discussion" section.

Response #7

This paragraph has been combined to Section 3.1 of "Results and discussion".

Comment #8

Page 21810, line 22: Authors should explain more what is observed in Fig. 2 (how the size, surface and volume distribution change in different seasons).

Response #8

One paragraph added:

"*In addition, the fitted GMDs of surface distribution were found to be 77.4 and 293nm for autumn, 22.1, 68.9 and 228 nm for winter, 77.4 and 253 nm for spring, and 12.9 and 268 nm for summer, respectively (not shown in the figures). In winter and summer seasons, one of the fitted surface GMDs was located at nucleation mode, showing the significant contribution of nucleation mode particles in these two seasons. Bimodal volume distribution was obtained for all seasons where the fitted volume GMDs were 96.3 and 372nm for autumn, 71.8 and 275 nm for winter, 99.5 and 339 nm for spring, and 99.5 and 237 nm for summer, respectively. The GMD of first volume mode was relatively stable in each season (i.e. 71.8-99.5 nm), but smaller GMD (237 nm) for the second volume mode was observed in summer. The results implied that a higher fraction of particles could have evolved from smaller size range (i.e. nucleation and Aitken modes) into accumulation mode, which coincided with our observation that NPF events occurred mostly in summer (see Section 3.4). Furthermore, this seasonal variability agrees with our previous findings that the growth rate of newly formed particles was correlated with the photolysis of ozone, an indicator of photochemical activity (Cheung et al., 2013). The causes responsible for the observed seasonal variations in PNCs will be detailed in the following sections.*"

Comment #9

Page 21811, line 1: Authors should refer here to the Table 1 where the ratios of concentration in different modes are presented, and also mention in the text what was the ratio for other seasons than summer.

Response #9

Page 21811, line 1: The corresponding discussion has been refereed to Table 1, and the ratio of $N_{4\text{-}25}/N_{4\text{-}736}$ for other seasons have been mentioned. The sentence of "It was revealed…" has been revised to "*It was revealed that the nucleation mode particles were predominant in the PNCs during autumn, winter and spring in the study area, whereas a distinct size distribution pattern was observed in summertime. In summer, the fraction of nucleation ($N_{4\text{-}25}$ / $N_{4\text{-}736}$) decreased to 0.44 (see Table 1) and the Aitken mode PNCs increased to be comparable to that of the nucleation mode, whereas the $N_{4\text{-}25}$ / $N_{4\text{-}736}$ ratios for other seasons ranged from 0.56 to 0.77 (see Table 1)*".

Comment #10

Page 21811, line 6: It is not entirely clear what the authors mean by writing "a large number of nucleation mode particles could have been shifted into the Aitken and/or accumulation modes". This should be explained in a more clear way.

Response #10

The confusing statement was removed and the paragraph was revised.

"*It was revealed that the nucleation mode particles were predominant in the PNCs during autumn, winter and spring in the study area, whereas a distinct size distribution pattern was observed in summertime. In summer, the fraction of nucleation ($N_{4\text{-}25}$ / $N_{4\text{-}736}$) decreased to 0.44 (see Table 1) and the Aitken mode PNCs increased to be comparable to that of the nucleation mode, whereas the $N_{4\text{-}25}$ / $N_{4\text{-}736}$ ratios for other seasons ranged from 0.56 to 0.77 (see Table 1). Observation from another aspect is that the PNC of nucleation mode ($N_{4\text{-}25}$) peaked in winter and reached a minimum in summer, whereas the PNCs of Aitken mode ($N_{25\text{-}100}$) and accumulation mode ($N_{100\text{-}736}$) reached their maxima in summertime. The changes in the size distribution in summer season were most likely due to the seasonally enhanced photochemical production of condensable vapors that, in turn, contributed to the growth of aerosol particles in the atmosphere.*"

Comment #11

Page 21811, line 20: If there are some previous studies where the mass of ultrafine particles has been measured in Asia, or in other conditions similar to the measurement site of this study, authors should refer also to those.

Response #11

The measurement of ambient mass concentration and chemical composition of UFPs is limited in Asia, although there were some studies measuring the UFPs mass concentration at roadside and tunnel. The only similar study reported was conducted in Taiwan which collected UFPs samples during May to November, but no seasonal mean value was reported in the study. Thus, we are unable to compare our data with other Asian data in the manuscript.

Comment #12

Page 21812, line 5: This sentence should be clarified, as it is rather difficult to understand.

Response #12

The reported organic carbon (OC) concentration as presented in this study, was not converted into organic matter (OM) which takes into account of hydrogen and oxygen of the organic compounds. Usually aged aerosols associated with higher oxygen and hydrogen in atmosphere, and thus characterized by a higher OM/OC ratio, while fresh aerosols have a lower OM/OC ratio. Thus, different conversion ratios, i.e. 1.2-1.6, have been applied to calculate OM by the OC depending on the environmental settings (i.e. rural or urban). Therefore if we assume all undefined portion is equivalent to the portion of OM after subtracting the OC fraction, a larger undefined portion of UFPs than PM1 suggested that the OM/OC ratio in UFPs was higher than that in PM1. This result implied that UFPs and PM_1 could be formed by different mechanisms. Corresponding discussion has been made in Section 3.2, Paragraph 2.

Page 21812, line 5: The sentence has been revised as "Thus a substantial amount of UFPs remained unidentified, which likely include hydrogen and oxygen associated with organic carbon (OC)."

Comment #13

Page 21812, line 16: This sentence should be revised as the annual average of PM1 is not actually presented in Fig. 3b.

Response #13

Page 21812, line 16: The first sentence has been revised as "*As shown in Fig. 3b, average PM_1 was estimated to be 14.7 $\mu g\ m^{-3}$ (seasonal means: 11.6-18.5 $\mu g\ m^{-3}$) in this study, which was similar to the results of a previous study in urban Taipei (average: 14.0 $\mu g\ m^{-3}$, Li et al., 2010).*"

Comment #14

Page 21813, line 19: Authors should refer here to Fig. 1 presenting the trajectories for each season.

Response #14

Page 21813, line 19: This sentence has been referred to Fig. 1.

"*The seasonal characteristics of PM_1 concentration and composition are attributed mostly to the changes in the origin areas of background air mass, which shifted from the Asia Continent to the western Pacific Ocean during summertime (see Fig. 1).*"

Comment #15

Page 21813, line 25: Authors should present in the manuscript (for example in this section) also figures showing the typical diurnal variation of size distributions in different seasons. For example, authors could make a surface plot (similar as in the bottom panel of Fig. 6) showing the median daily variation of particle size distribution for each season.

Response #15

Discussion on the diurnal variation in different seasons has been added.

Page 21814, line 7: The following sentences have been inserted after "…in other seasons.":

"*In addition, the diurnal variation of particle size distribution (see Figure 5) provided further information about the variations in PSD. Two nucleation bursts were distinctly observed in morning and afternoon traffic peak hours in autumn, winter, and spring, while a typical PSD pattern of nucleation event (a banana curve) was dominant in summer. This result is as expected because the photochemical production of nucleation mode particles is more intense during warm seasons (Cheung et al., 2011). Moreover, as discussed in previous section, the photochemical reactions could produce condensable organics that allows the newly formed nucleation mode particles to grow into the Aitken mode. The relatively small differences between the daytime and nighttime $N_{4\text{-}736}$ in autumn and winter indicated that the photochemical contribution in PNCs was declined as compared to that in summertime.*"

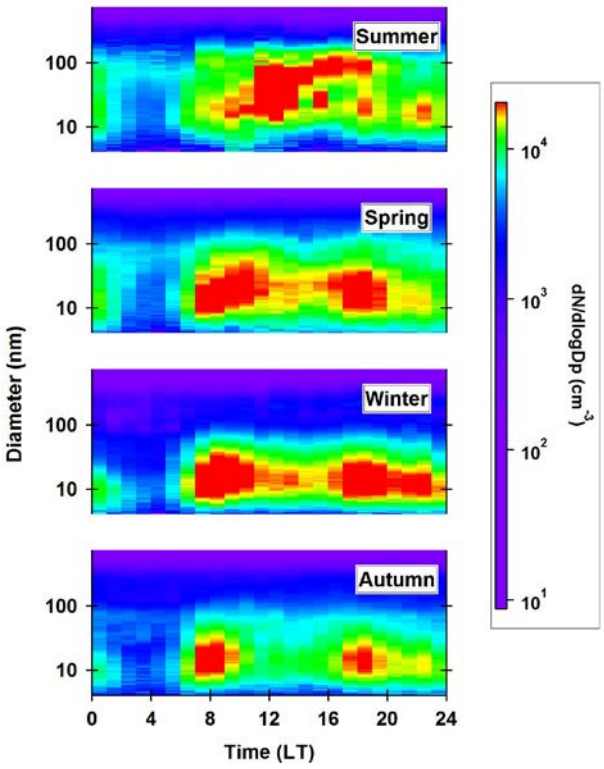


Figure 5. Diurnal variation of particle number size distribution in each season. From lower panel to top panel: autumn, winter, spring and summer.

Comment #16

Page 21814, line 3: Authors utilize the difference between particle number concentrations during day and night to show if there is formation of new particles taking place in different seasons. However, it is not clear how other factors (e.g. boundary layer dynamics) affect the seasonal variation of that difference.

Response #16

The impact of boundary layer dynamics affects both daytime and nighttime data within the same season. For example, the boundary layer height should be lower in winter and this both occurs in daytime or nighttime; similarly, a higher boundary layer favors the dispersion process in summertime, this both occurs during daytime or nighttime. Thus, the boundary layer effect on each season should not have a major impact on the calculated difference on particle number concentration between daytime and nighttime in different seasons.

Comment #17

Page 21814 line 23: When calculating correlation coefficients between particle number concentrations and NO_x, it would be better to first take logarithm of both variables and then calculate the correlation coefficients. Otherwise single data points

can have too large effect on the value of the correlation coefficient.

Response #17

The correlation coefficients between particle number concentrations and NO_x have been re-calculated, and the results were compared to previous calculation as below:

	Winter		**Summer**	
	New (taking logarithm)	**Old**	**New (taking logarithm)**	**Old**
$N_{4\text{-}25}$	0.84	0.71	0.81	0.81
$N_{25\text{-}100}$	0.88	0.87	0.83	0.83
$N_{100\text{-}736}$	0.31	0.29	0.72	0.61

The correlation coefficients calculated after taking logarithm of both variables were slightly improved, and but it did not change the conclusion drawn on the relationship between PNC and NO_x as the values were similar. However, the correlation coefficients were calculated based on the values without taking logarithm in previous studies, and for comparison with previous results, we would like to keep the original calculation method.

Comment #18

Page 21815, line 10: Some numbers for the slope values should be given also in the text, not only in the figure.

Response #18

Page 21815, line 10: These sentences "The slope values…on particle concentration." have been revised to "*The slope values can serve as a relative emission factor of particles per NOx, which indicates the degree of influence of vehicle emission on the PNCs (Cheung et al., 2013). The corresponding slope values for $N_{4\text{-}25}$, $N_{25\text{-}100}$, and $N_{100\text{-}736}$, were found to be 279, 163, 18 $cm^{-3}\cdot ppb^{-1}$ in winter, and 239, 330, 155 $cm^{-3}\cdot ppb^{-1}$ in summer. Larger sum of slope values (724 vs.460 $cm^{-3}\cdot ppb^{-1}$) was found in summertime compared to winter period, evidencing a greater influence of the vehicle emission on particle number concentration.*"

Comment #19

Page 21815, line 13: It is not entirely clear what is meant by "demonstrate the size shift effects of particle growth", so the sentence should be written in a more clear way.

Response #19

Page 21815, line 13: We agreed that the sentence "Furthermore, the lower…" is confusing and thereby have deleted it.

Comment #20

Page 21816, line 5: Based on Fig. 6 the increase of PM10 from 10 to ~100 μg m^{-3} is at least partly related to diurnal variation, and not only to the change in the wind direction (after the change in wind direction PM10 is still for some hours clearly above 10 μg m^{-3}).

Response #20

Thank you for your comment on PM_{10} variation. We agreed that the variation of PM_{10} and other pollutants are affected both by regional and local effects. The corresponding sentence has been revised to clarify the discussion.

"*It should be noted that the variations of measured pollutants were not solely influenced by the long-range transport, but also partly due to the variation of local pollution and boundary dynamics.*"

Comment #21

Page 21816, line 12: Authors should mention if there were any new particle formation events during LRT events.

Response #21

On 7 April 2013, a LRT event was concurrently occurred with a dust event, and a banana shape of particle number size distribution was observed. However, the dominating diameter measured at the initial stage of this event was around 40-50 nm. This observation could due to the NPF process occurred in upwind area. Recent study by Nie et al. (2014) proposed a new particle formation mechanism associated with dust particles. The particle precursors attached on the dust surface will under heterogeneous reaction to form particles, then the secondary particles will released from the dust surface to the atmosphere during the transport of air masses. Due to the unique factors (including the impact of dust storm and other anthropogenic pollutants), we differentiated this case from the NPF events, and a discussion was added in the manuscript to address this special event.

"*The result suggested that the influences of local vehicle emission on PNCs were still in place, whereas growth of particles due to secondary production of condensable vapors could have been suppressed, as NPF was rarely observed during the LRT events. It is noteworthy that a weak dust transport event was observed on 7 April 2013 where a banana shape was depicted in the PSD, evidencing that secondary formation of particles could have had occurred. However, the dominating diameter of particles was ~40-50 nm at the initial stage of the event. The banana shape of PSD data was initiated since ~06:00 LT until 21:00 LT, when the northeasterly wind prevailed. The*

PM_{10} and O_3 also increased from minima of 44 μg m^{-3} (at 06:00 LT) and 25 ppb (at 05:00 LT) to the daily maxima of 92 μg m^{-3} (at 17:00 LT) and 61 ppb (at 16:00 LT). This result showed that the NPF process could have occurred in the upwind area where newly formed particles were transported to the study site, or heterogeneously formed particles were released from the dust surface during the long-range transport of air pollutants (Nie et al., 2014)."

Comment #22

Page 21816, line 20: Authors should mention if the value given for the wind speed is mean or median.

Response #22

"…lower wind speed…" has been revised to "…*low average wind speed.*…".

Comment #23

Page 21817, line 1: This section should be in a different place, not in the end of the "Results and discussion" section but closer to the beginning, as the observation of the frequency of NPF events helps to understand also other results (e.g. the seasonal variation in the composition of particles).

Response #23

The order of the discussion has been rearranged.

Section 3.6 has been moved to Section 3.4. Corresponding figure and table numbers have been revised in the manuscript.

Comment #24

Page 21817, line 4: Figure 8 should be combined with Fig. 5 and discussed in the same section. Authors should first tell how often they observed NPF events during different seasons and then use the correlation with NO_x only to this observation.

Response #24

The discussion on NPF (Section 3.6) has been moved to Section 3.4, and we first discussed the frequency of the NPF events different seasons. Figure 8 shows the influence of secondary sources on PNC, and Figure 5 shows the influence of primary sources. As there will be too many data shown if the two figures were combined, we would like to discussion the influences of these two sources separately.

The corresponding discussion on NPF events (i.e. first paragraph of Section 3.4 in revised manuscript) has been rewritten as below:

"*As shown in previous study, the NPF events were frequently observed in summer,*

which subsequently induced a notable increase in N_{4-25} in urban Taipei (Cheung et al. 2013). The frequency of NPF events was found to be 10 out of 84 measurement days and the events were observed in autumn (1 out 23 days), spring (3 out of 26 days) and summer (6 out of 14 days) seasons. Figure 6 (a-d) shows the scatter plots of N_{4-25} against NOx for daytimes in each season. During the NPF events, a non-linear relationship between these two parameters was usually observed during the daytime (Cheung et al. 2013). The results showed that clear NPF events were observed often in summer and occasionally in spring, but rarely in autumn and winter in the study area. The averaged particle growth and formation rates were found to be 4.0±1.1 nm h^{-1} and 1.4±0.8 cm^{-3} s^{-1}, which were comparable to those measured in other urban studies in Asian countries such as Hong Kong (average: 6.7 nm h^{-1}, Wang et al., 2014) and Beijing (average: 5.2 nm h^{-1}, Wang et al., 2013). The particle growth and formation rates of each case are listed in Table 2."

Comment #25

Page 21817, line 12: Also some other references on particle growth rates could be mentioned here.

Response #25

Other references on particle growth rates have been mentioned.

The sentence "The averaged particle…" has been revised to "*The averaged particle growth and formation rates were found to be 4.0±1.1 nm h^{-1} and 1.4±0.8 cm^{-3} s^{-1}, which were comparable to those measured in other urban studies in Asian countries such as Hong Kong (average: 6.7 nm h^{-1}, Wang et al., 2014) and Beijing (average: 5.2 nm h^{-1}, Wang et al., 2013)*".

The following studies have been added to the references list:

Wang, D., Guo, H., Cheung, K. and Gan, F. (2014). Observation of nucleation mode particle burst and new particle formation events at an urban site in Hong Kong. Atmospheric Environment, 99, 196-205.

Wang Z. B., Hu, M., Suu, J.Y., Wu, Z.J., Yue, D.L., Shen, X.J., Zhang, Y.M., Pei, X.Y., Cheng, Y.F. and Wiedensohler, A. (2013). Characteristics of regional new particle formation in urban and regional background environments in the North China Plain. Atmospheric Chemistry and Physics, 13, 12495-12506.

Comment #26

Page 21817, line 14: Table S3 should not be in the supplementary but in the main text because the information on growth rates and formation rates is relevant.

Response #26

Table S3 has been included in the main text.

Page 21817, line 14: The sentence has been revised as "*The particle growth and formation rates of each cases are listed in Table 2.*"

Comment #27

Page 21817, line 18: Authors should show that low PM10 and high sulfuric acid proxy favors new particle formation for example by studying their correlation with $N_{4\text{-}25}$, or then by showing their median diurnal variation for days with NPF events and days without NPF events. Showing only median of these variables for different seasons is not enough for drawing conclusions about their importance for new particle formation. Authors could also study the effect of the air mass origin on the occurrence of NPF events by studying the air mass trajectories. In addition, authors should refer to some of the earlier studies where low condensation sink and high sulfuric acid concentration have been observed to favor NPF events.

Response #27

The observed $N_{4\text{-}25}$ not only affected by the secondary production of particles, also associated with the local vehicle emission in this study. To examine the influence of SO_2 and condensation sink (CS) on NPF, the scatter plot between the UVB*SO_2 against CS has been deployed. This method has been applied in previous studies (e.g. Gao et al., 2009; Nie et al. 2014) to demonstrate the effect of sulfuric acid and pre-existing particles on NPF.

In this study, the spatial resolution of applied meteorological data is 1° x 1° (around 100km x 100km), the resolution of meteorological data is not sufficient for an accurate back-trajectories analysis on a local scale as in our study site. Nevertheless, a back-trajectory drawn during a NPF event will only indicate the pathway of the air mass, but it could be originated from any points along the path. Hence, a detailed emission inventory (which is not available for UFP) and back trajectory calculations are needed which required a further analysis and is not within the scope of the current study.

The second paragraph has been rewritten as below:

"*Table 3 summarizes the averages of $N_{4\text{-}25}$, PM_{10}, H_2SO_4 proxy (as UVB*SO_2/condensation sink) and wind speed for each season. The dominating factors associated to the frequent particle formation in summertime were the low PM_{10} concentration (35.6 µg m^{-3}) and high H_2SO_4 proxy (493.1 ppb W m^{-2} s). The association of sulfuric acid production and the NPF events agreed with the elevated*

mass concentration of sulfate in UFPs during summertime (shown in Table S1), as well as the results of previous urban studies (Woo et al. 2001; Cheung et al. 2013). This strongly suggested that the new particle formation was mainly driven by the photochemical oxidation of SO_2 under low condensation sink conditions (Nie et al., 2014), where the SO_2 could be transported from the upwind area on the summer monsoons (see Figure 1d). Contrarily, the absence of particle formation events in wintertime could be attributed to the declined photochemical production of H_2SO_4 as well as suppression of NPF by particles transported from the Asian continent (Lin et al., 2004). The results of this work evidenced that low PM_{10} concentration and high sulfuric acid production favored the particle formation process in urban areas."

In the manuscript, earlier studies on the favorable condition for NPF events have been referred.

"This strongly supports…" *has been revised to "This strongly suggested that the new particle formation was mainly driven by the photochemical oxidation of SO_2 under low condensation sink conditions (Gao et al., 2009; Nie et al., 2014), where the SO_2 could be transported from the upwind area on the summer monsoons (see Figure 1d).*"

The reference below has been added:

Gao, J., Wang, T., Zhou, X. Wu, W. and Wang, W. (2009). Measurement of aerosol number size distributions in the Yangtze River delta in China: Formation and growth of particles under polluted conditions. Atmos. Environ., 43, 829-836.

Nie, W., Ding, A., Wang, T., Kerminen, V.-M., George, C., Xue, L., Wang, W., Zhang, Q., Petäjä, T., Qi, X., Gao, Xiaomei, Wang, X., Yang, X., Fu, C. and Kulmala. (2014). Polluted dust promotes new particle formation and growth. Scientific Reports, 4, 6634.

Comment #28

Page 21818, line 24: It should be again explained in a more clear way what is meant by "shifting of the nucleation mode particles".

Response #28

The statement was relevant to the growth of particles from condensation mode range to Aitken mode size range. The sentence has been revised.

"It was revealed from the measurements of PSD that a large number of nucleation mode particles could have evolved into the Aitken mode during summertime, which was most likely relevant to the photochemical production of condensable vapors that, in turn, could have contributed to the growth of particles in the atmosphere".

Comment #29

Page 21827, Table 3: Condensation sink should be added to the table. In addition, the median values UVB and SO_2 should be shown separately instead of showing their product.

Response #29

The individual values of the condensation sink, UVB and SO_2 have been included in Table 3.

Comment #30

Page 21830, Figure 3: It should be mentioned in the caption if the values are seasonal averages or medians.

Response #30

Page 21830, Figure 3: The figure caption has been revised to "*Seasonal average concentration and composition of **(a)** ultra-fine (UFPs) and **(b)** sub-micron (PM1) particles observed at the TARO in Taipei, Taiwan from autumn 2012 to summer 2013.*"

Comment #31

Page 21832, Figure 5: It should be explained if the data points in the figure are averages for a certain time interval.

Response #31

In Section 2.2, it was already mentioned that the original PNCs data time resolution was 5mins, and the hourly average data were calculated for data analysis. To make it clearer, the use of hourly average data is mentioned in the caption of Figure 5 as below:

"***Figure 5.** Scatter plots for hourly averaged PNCs vs. NO_x measured during the time period of 20:00-04:00 (LT) in **(a)** winter and **(b)** summer, with classification of various particle size ranges.*".

Comment #32

Supplement, Table S3: This table should be moved to the main text. It should be explained in the methods section how the occurrence of NPF events was determined, and how growth rates and formation rates were calculated.

Response #32

Table S3 has been moved to the main text. The discussions on the classification of NPF events and calculation of growth rates and formation rates have been added to Section 2 of methodology.

Section 2.3 Classification of new particle formation and calculation of the particle growth and formation rates

A NPF event is defined as the increase of the number concentration of nucleation mode particles, where those particles are growing into Aitken and/or accumulation mode size range (≥ 25 nm) and last for a few hours until they disappear into the atmosphere by condensation/ coagulation sinks (Dal Maso et al., 2005). The calculation of particle growth rate (GR) was represented by the rate of geometric median diameter (GMD) changes during the period of nucleation mode particles growing through 25 nm (Cheung et al., 2013). The formation rate (J) of nucleation mode particles for each NPF event was calculated for the particle size ranging from 4-25 nm according to the method of Dal Maso et al. (2005). Formation rate is defined as the sum of the apparent formation rate (dN_{4-25}/dt) and the coagulation loss rate during the NPF event. It should be noted that the reported apparent particle formation rate is expected to be smaller than the actual nucleation rate, since some fractions of formed nuclei are always scavenged by coagulation into larger pre-existing particles before they grow larger by condensation (Lehtinen et al., 2007).

The following references have been added to the reference list:

Lehtinen, K.E.J., Dal Maso, M., Kulmala, M. and Kerminen, V.-M. (2007). 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, doi:10.1016/j.jaerosci.2007.06.009.

Dal Maso, M., Kulmala, M., Riipinen, I., Wagner, R., Hussein, T., Aalto, P.P. and Lehtinen, K.E.J. (2005). Formation and growth of fresh atmospheric aerosols: eight years of aerosol size distribution data from SMEAR II, Hyytiälä, Finland. Boreal Environ. Res., 10, 323-336.

Technical corrections

Comment #33

Page 21806, line 7: The abbreviation (PNC) should be explained here.

Response #33

Page 21806, line 7: "…nucleation mode PNCs…" has been revised to "*nucleation mode particle number concentrations*".

Comment #34

Page 21806, line 24: The sentences starting with "To attain a better understanding …" is too long.

Response #34

Page 21806, line 24: The sentence "To attain a…" has been revised to "*A 1-year aerosol characterization experiment was conducted in urban area of Taipei, Taiwan. The aim of this study is to attain a better understanding of the seasonal variations of ultrafine and sub-micron particles and the factors affecting particle formation, particularly under the influences of Asian monsoon circulations.*"

Comment #35

Page 21808, line 20: Use of lash (/) here might be confusing for the reader.

Response #35

Page 21808, line 20: The sentence has been revised to "*…and a total of 69 and 75 sets of UFPs and PM_1 samples were collected during the entire investigation period (sample sets collected in autumn, winter, spring and summer were 20, 15, 25, and 9 sets for UFPs, and 21, 16, 25, and 13 sets for PM_1, respectively).*".

Comment #36

Page 21810, line 12: It would be better write "particle size distributions" than use the abbreviation.

Response #36

Page 21810, line 12: "3.1 PNCs and PSDs in respective seasons" has been revised to "*Particle number concentration and size distributions in respective seasons*".

Comment #37

Page 21810, line 13: The abbreviation (PNC) should be explained here.

Response #37

Page 21810, line 13: "PNCs" has been revised to "*particle number concentrations*".

Comment #38

Page 21810, line 15: The concentrations are written here in the wrong order (first should be the concentration in spring and then the concentration in winter).

Response #38

Page 21810, line 15: "Relatively higher…" has been revised "*Relatively higher $N_{4\text{-}736}$ were observed in spring and winter with median concentrations of $19.4x10^3$ and $17.4x10^3$ cm^{-3}, respectively, followed by summer ($16.6x10^3$ cm^{-3}) and were minimum in autumn ($13.9x10^3$ cm^{-3}).*".

Comment #39

Page 21810, line 24: The sentence starting with “It was relieved…” is too long.

Response #39

Page 21810, line 24: The sentence “It was relieved…” has been revised to “*It was revealed that the nucleation mode particles were predominant in the PNCs during autumn, winter and spring in the study area, whereas a distinct size distribution pattern was observed in summertime. In summer, the fraction of nucleation ($N_{4\text{-}25}$ / $N_{4\text{-}736}$) decreased to 0.44 (see Table 1) and the Aitken mode PNCs increased to be comparable to that of the nucleation mode, whereas the $N_{4\text{-}25}$ / $N_{4\text{-}736}$ ratios for other seasons ranged from 0.56 to 0.77 (see Table 1)*”.

Comment #40

Page 21811, line 11: It seems that “dominated” is not necessarily the correct choice of word here. May be “correlated” could be a better word.

Response #40

Page 21811, line 11: The word “dominated” has been revised to “*correlated*”.

Comment #41

Page 21811, line 22: The sentence starting with “For the chemical composition…” is too long.

Response #41

Page 21811, line 22: The sentence “For the chemical composition…” has been revised to “*For the chemical composition, organic carbon (OC) was found to be the major mass contributor, which accounted for 29.8 % (seasonal means ranging from 26.9 to 33.4 % for various seasons) of averaged mass concentration of UFPs. Elemental Carbon (EC) was the second major component with averaged mass contribution of 5.1 % (seasonal means: 2.4–7.6 %), followed by sulfate (SO_4^{2-}) at 4.3 % (seasonal means: 3.4-6.4%) and nitrite (NO_2^-) at 2.9% (seasonal means: 0.9-7.3%).*”

Comment #42

Page 21812, line 24: “UPFs” should be “UFPs”.

Response #42

Page 21812, line 24: “*UPFs*” has been revised to “*UFPs*”.

Comment #43

Page 21813, line 12: “Maximal” and “minimal” should be “maximum” and “minimum”.

Response #43

Page 21813, line 12: "*Maximal*" and "*minimal*" have been revised to "*maximum*" and "*minimum*".

Comment #44

Page 21815, line 2: Use of lash (/) here is confusing.

Response #44

Page 21815, line 2: The sentence "The robust…" has been revised to "*The robust correlation of NO_x and N_{4-25}, also NO_x and N_{25-100} suggested that local vehicle emission is the predominant source of UFPs throughout a year.*".

Comment #45

Page 21816, line 10: Writing "particles stayed at a low level" would be better.

Response #45

Page 21816, line 10: "kept at a low level" has been revised to "*remained at a low level*".

Comment #46

Page 21817, line 7: Instead of "remarkable NPF events" it could be better write e.g. "clear" or "strong".

Response #46

Page 21817, line 7: "remarkable NPF events" has been revised to "*clear NPF events*".

Comment #47

Page 21818, line 2: The abbreviations should be explained again here.

Response #47

Page 21818, line 2: The sentence "The mass…" has been revised to "*The mass concentration and chemical composition of ultrafine particles (UFPs) and submicron particles (i.e. PM_1) as well as the particle number concentration (PNCs) and size distributions (PSDs) with size ranging from 4 to 736 nm were measured during four seasonal campaigns in the period from October 2012 to August 2013 at the TARO, a subtropical urban aerosol station in Taipei, Taiwan.*"

Comment #48

Page 21825, Table 1: It should be explained in the caption that the ratios between the concentrations in different modes are presented in the last columns.

Response #48

Page 21825, Table 1: The sentence "*The fractions of N_{4-25} and N_{4-100} to total PNCs*

were presented in the last two columns." has been amended in the caption.

Comment #49

Page 21828, Figure 1: Figure is too small; it is not possible to read the text in the labels on the right panel. It should be explained in the figure caption what the different colors in wind roses present. In addition, most of the other figures in the manuscript are also too small and include text with too small font and/or too thin lines.

Response #49

Page 21828, Figure 1: The figure caption has been revised to explain the color code for wind rose plots. Other figures in the manuscript have been revised with larger fonts and thicker lines as suggested.

Comment #50

Page 21831, Figure 4: Background of the figures should be white. The same applies to Fig. 8.

Response #50

Background colors of Figures 4 and 8 have been revised to white color.

Comment #51

Page 21833, Figure 6: The color bar should be move to the bottom of the figure. The color scale could be shown in logarithmic scale instead of linear scale.

Response #51

Figure 6 has been modified by moving the color bar to right hand side and is shown on a logarithmic scale.

Referee #2

This manuscript reported the measurements of submicron and ultrafine particles in four different seasons in an urban area in Taiwan, and provided useful information on particle number size distributions, chemical compositions, source apportionment, and new particle formation events. The manuscript is overall well written and fits the scope of ACP. But the sampling periods, totally 84 days, were quite short in each season. Their representative for the whole season needs to be verified. I recommend some revisions before this manuscript can be published on ACP.

Specific comment

Comment #1

The representative of sampling periods for the whole seasons needed to be verified. In some seasons, the campaign was only conducted for about 2 weeks, which make it doubtful for the statistical significance of the results. Therefore, detailed information about the meteorology and pollution parameters is needed. E.g. Page 21811, line 11, the exact ozone concentrations are needed to define the strength of photochemical processes.

Response #1

We agreed that the relatively short of sampling periods during the whole year is not significant to discuss the seasonality of measured parameters (i.e. PNC, PSD) in the study area. However, the back-trajectories of the sampling days as shown in Figure 1 indicated that both winter and summer monsoon circulation patterns, which are the major characteristics of the wind circulation pattern in East Asia region, have been captured in this study. Therefore, the variations of PNC/PSD should be representative to the seasonal variations of those measured parameters in this study. Nevertheless, some events (certain dust storms events) could not be entirely captured in certain periods when measurements were not conducted, we thus define our scope to study the "seasonal variations" which are clearly demonstrated by our results. As such, we revised the manuscript title to "*Seasonal variations of ultrafine and sub-micron aerosols in Taipei, Taiwan: implications for particle formation processes in urban areas*" for more accurate description of this study.

Comment #2

I suggest reconsidering the title of the manuscript.

Response #2

The title of the manuscript has been revised to "*Seasonal variations of ultrafine and sub-micron aerosols in Taipei, Taiwan: implications for particle formation processes in urban areas*" to better describe this study.

Comment #3

Define the “long range transport (LRT)” to distinguish LRT and Non-LRT.

Response #3

The definition and classification scheme of LRT has been discussed in Section 3.5.

Comment #4

Page 21804, line 6: define TARO

Response #4

Page 21804, line 6: Definition of TARO has been added.

“…at the TARO (Taipei Aerosol and Radiation Observatory),…”

Comment #5

Page 21804, line 9: particle size distribution is not an accurate definition. Please clarify it as particle number size distribution, or particle mass size distribution.

Response #5

Page 21804, “size distribution (PSD)” has been revised to “*number size distribution (PSD)*”.

Comment #6

Page 21804, line 11: change “highest” to “lowest”.

Response #6

The highest UFPs concentration was obtained in summer. Thus, no revision is needed.

Comment #7

Page 21805, line 4-5: there was not enough evidence to support this conclusion.

Response #7

We agreed with the comments and revised the statement as

“The results of this study implied the significance of secondary organic aerosols in the seasonal variations of UFPs and the influences of continental pollution outbreaks in the downwind areas of Asian outflows.”

Comment #8

Page 21808, line 6: provide the detailed information of the size cutoff for each stage of MOUDI, and the sampling flow.

Response #8

We only use one stage of the MOUDI impactor to collect UFPs (100nm), and the PM_1 samples were collected by PQ-200 (BGI Inc.) sampler. Thus, the size-cut of other stages (of larger sizes) was not stated in this study as they were not used. Nevertheless,

the design paper of MOUDI (i.e. *Marple et al., 1991*) was cited where the features of MOUDI are described in details.

The sampling flow rates of MOUDI and PQ-200 samplers were added.

Page 21808, line 6: The sentence of "*The sampling flow rate of MOUDI sampler was 30 lpm.*" was inserted after the sentence "…a collector of UFPs.".

Page 21808, line 9: The sentence "Besides, a pair…" has been revised to "*Besides, a pair of PM_1 samplers, each consisted of a standard aerosol sampler (PQ-200, BGI Inc.) and a PM_1 sharp cut cyclone, were deployed to collect PM_1 samples, with 16.7 lpm sampling flow rate.*".

Comment #9

Page 21808, line 21: detailed sampling periods of MOUDI are needed.

Response #9

The sampling period of MOUDI was already mentioned in Page 21808, line 21. The corresponding sentence has been revised for better clarity.

"*The sampling duration of each sample set (for both MOUDI and PQ-200 samplers) was from 14:00 – 12:00 LT (22 hr)…*".

Comment #10

Page 21808, line 25-26: at least one reference is needed to describe the instruments of PM10, NOx, SO2 and O3.

Response #10

The reference for trace gas instruments has been added.

"*The details of instrumentation setup for trace gases measurement are referred to Cheung et al. (2013).*".

Comment #11

Check and unify the effective digital for the all manuscript.

Response #11

The effective digital of the measured parameters (i.e. PNC, PM) have been unified.

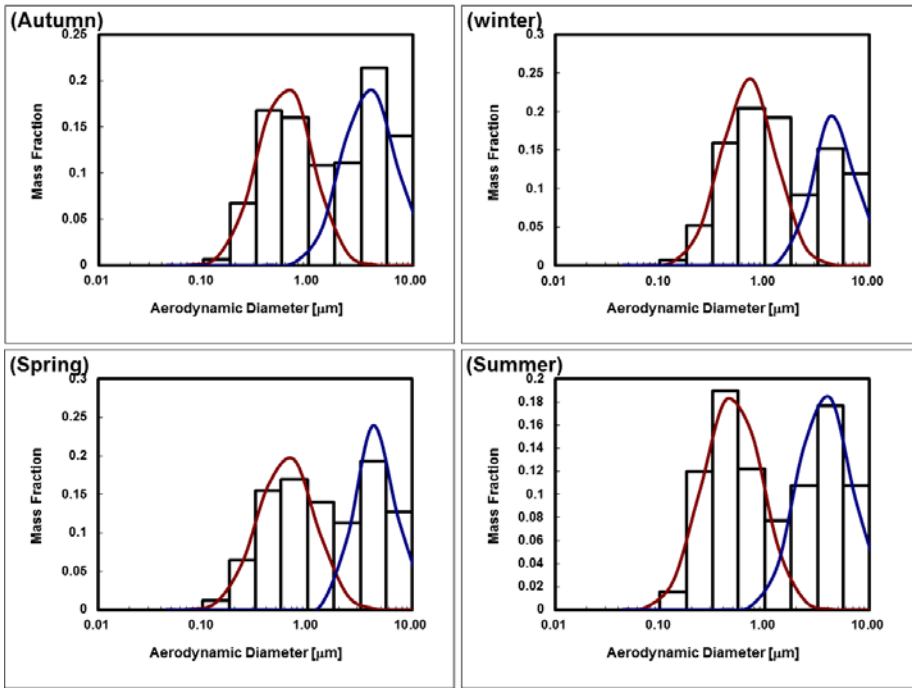
Comment #12

Section 3.2: It will be good to compare the particle volume size distribution (SMPS) and particle mass concentration (MOUDI).

Response #12

Thanks to the comments. Indeed this analysis is undergoing. Following plots illustrate the averaged mass size distribution for each season. Typical bimodal distribution is shown. Among the major findings is that the mode diameter of fine (accumulation) mode particles in summer was significantly smaller than in other seasons. This is well consistent with the seasonal pattern of the "second volume mode" retrieved from SMPS measurements.

Because the inter-comparison of size distribution is not within the scope of this paper, we prefer publishing this result in open discussion but not including into the manuscript. We shall present analysis of the size distribution of respective particulate species in a separate paper.



Comment #13

Page 21815, line 1: what is the reason of the high correlation of NOx and accumulation mode particles in summer? NOx is a tracer for primary vehicle emission, but accumulation mode particles are aged particles and rarely are emitted directly from vehicles.

Response #13

In contrast to the winter case, the Asian pollution outbreaks did not occurred in summer. Thus the PNC of accumulation particles was also dominated by local pollution, in particular vehicular exhausts. The PNC-NOx correlation evidenced this attribution. The contribution could be direct emission of particles or emitting

precursors of secondary aerosols. The following sentences were added to address this.

"*Interestingly, moderate correlation between the PNC of accumulation mode particles ($N_{100-736}$) and NOx was also observed in summer. Given that the Asian outflows were ceased during summertime, this correlation evidenced substantial contribution of local sources, particularly vehicular emissions, to the PNC of accumulation mode particles in Taipei, Taiwan.*"

Comment #14

Page P21815, line 8: the figure 6 should be figure 5

Response #14

Page P21815, line 8: The corresponding sentence has been revised.

Comment #15

The author attitude the large difference between observed PNCS in daytime and nighttime to the more intense photochemical production of particles. However, pattern of particle size distribution in summer nighttime with weak photochemical activity was also obviously different to that in the nighttime of other seasons. Are there other reasons for the special difference between observed PNCS in daytime and nighttime?

Response #15

The method used in this study to calculate the influence between photochemical production of PNCS in daytime and nighttime by assuming the photochemical activities are the major contributor to new particle formation process. We agreed that there are other sources influencing the PNCs in urban environment such as condensation of vapors and coagulation among the newly formed particles and primary emitted particles. However, these processes will not result in the difference in daytime and nighttime PNCs as observed in this study. We think that photochemical reaction is the still the major attributing factor to the difference between the PNCs in daytime and nighttime.

The particle size distribution in summer nighttime depicted two peaks around the nucleation mode and Aitken mode. For other seasons, a different pattern is depicted where there is a clear peak around nucleation mode, but a smaller Aitken mode also exists although it is at a smaller magnitude (indicated by the blue arrow). The observation showed that a large Aitken mode exist during the summer nighttime. Although no photochemical reaction occurred during the nighttime, a portion of newly formed particles during the summer daytime could still remain as the background concentration around the Aitken mode.

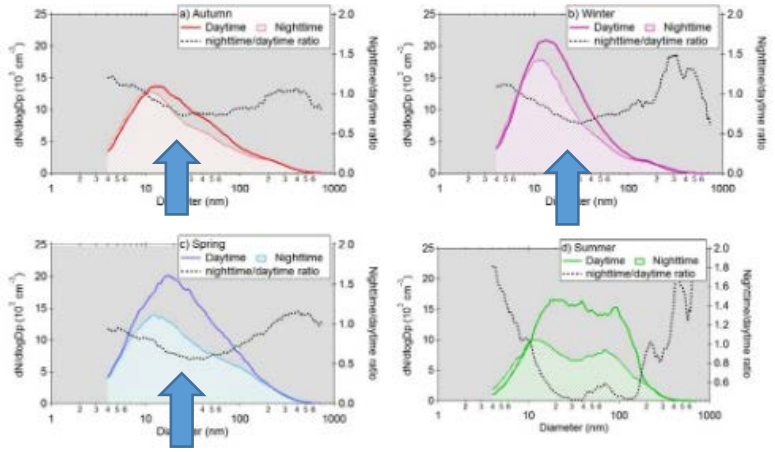


Figure 4. Median PSDs measured during the daytime (07:00–17:00 LT) and nighttime (17:00–07:00) in **(a)** autumn, **(b)** winter, **(c)** spring and **(d)** summer.

The discussion about the influence of other possible sources on PNCs has been added in the manuscript.

"In urban environment, the possible sources influencing the PNC and PSD are complicated which not only include the direct emission from primary sources, but also interaction between the newly formed particles, pre-existing particles and condensing vapors by the condensation and coagulation processes. Nevertheless, these processes occurred throughout the day and will not result in the difference in daytime and nighttime PNCs as observed in this study. We think that photochemical reaction is the still the major attributing factor to the difference observed between the daytime and nighttime PNCs."

Comment #16

Condensation sink is an important parameter for new particle formation. It should be calculated and provided in Table 3.

Response #16

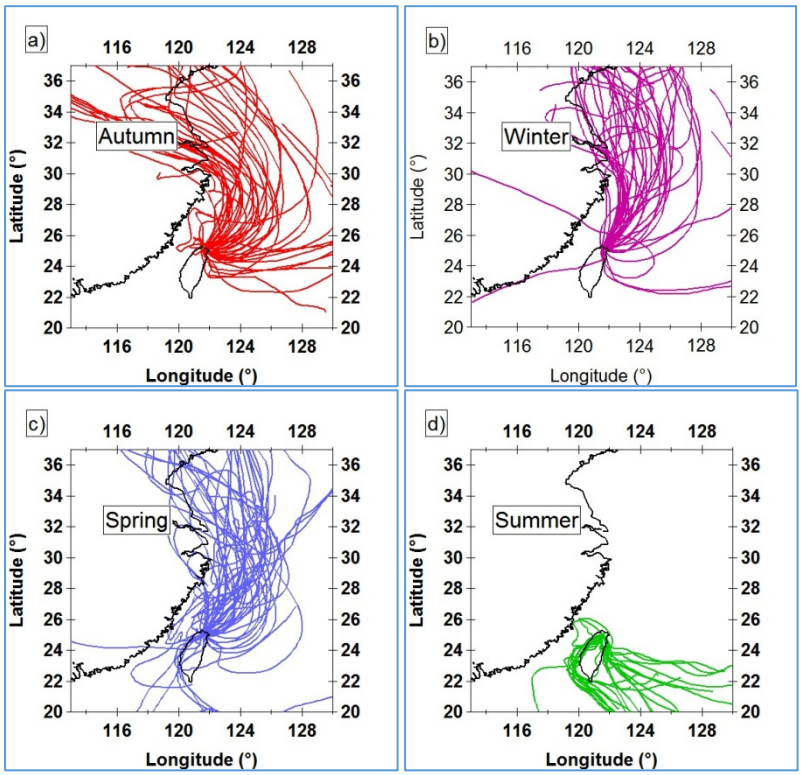
The individual values of the condensation sink, UVB and SO2 have been included in Table 3.

Comment #17

Figure 1: Given the influence of the regional transport from mainland China, it's better to add the map of southeast China in the figure.

Response #17

The southeast China region has been included in the map (Figure 1).



Comment #18

Figure 3: Due to the Asian monsoon, it is generally accepted that the air pollution is more serious in winter than that in summer in Taiwan and Hong Kong. Can the authors explain why the max PM concentration appeared in summer? Was it identical with other studies?

Response #18

Up to date, there is no other similar urban study on seasonal variation of UFPs mass and chemical composition in Hong Kong and Taiwan. In this study, higher PM_1 concentrations were obtained during spring and winter when long-range transport was dominant. For UFPs, the highest value was obtained in summer, and this suggested that the formation mechanisms of PM_1 and UPFs were different. A UFPs source apportionment study was conducted in urban Taipei during May to November 2011 (Gugamsetty et al. 2012) which indicated that the contribution by secondary aerosols were higher in $PM_{0.1}$ than that for $PM_{2.5}$ and PM_{10}. This difference of formation mechanisms has been discussed in Section 3.2.