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.*"

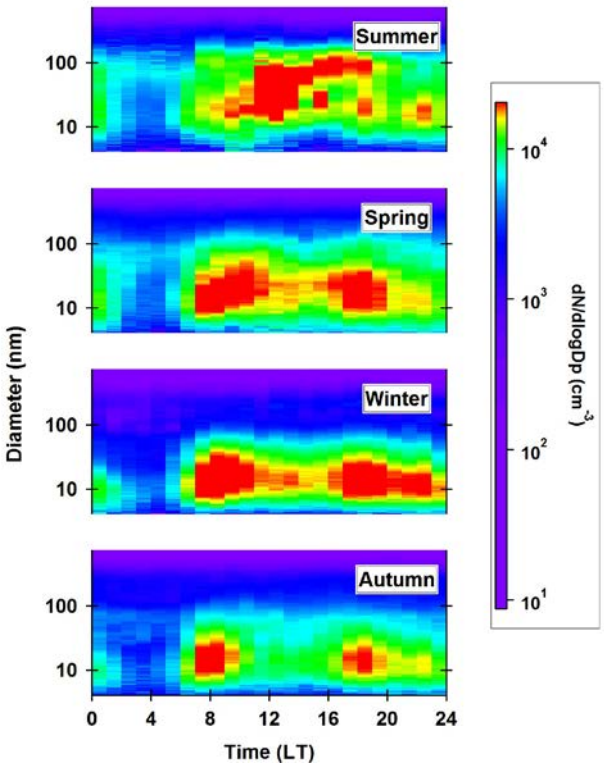


Figure 5. Diurnal variation of particle number size distribution in each season. From lower panel to top panel: autumn, winter, spring and summer.

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.

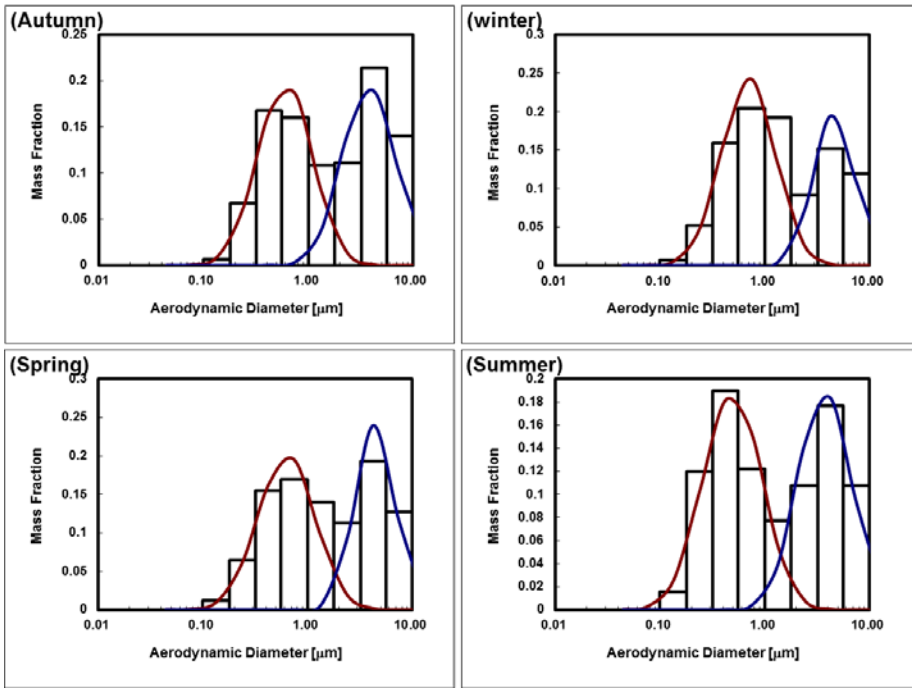
Comment #12

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.

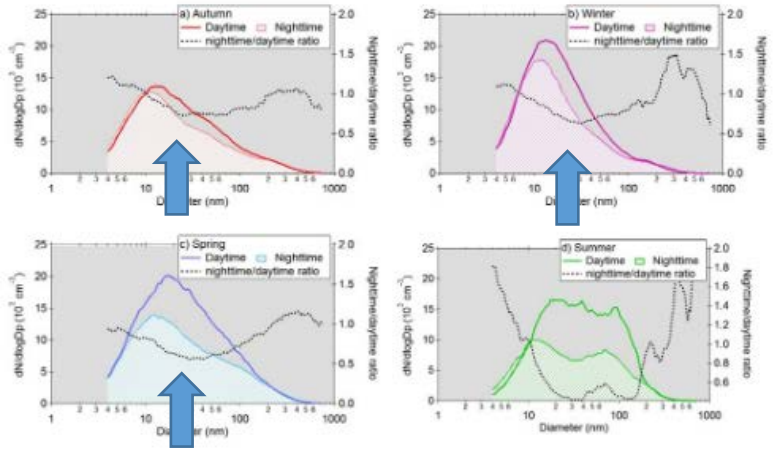


Figure 4. Median PSDs measured during the daytime (07:00–17:00 LT) and nighttime (17:00–07:00) in **(a)** autumn, **(b)** winter, **(c)** spring and **(d)** summer.

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

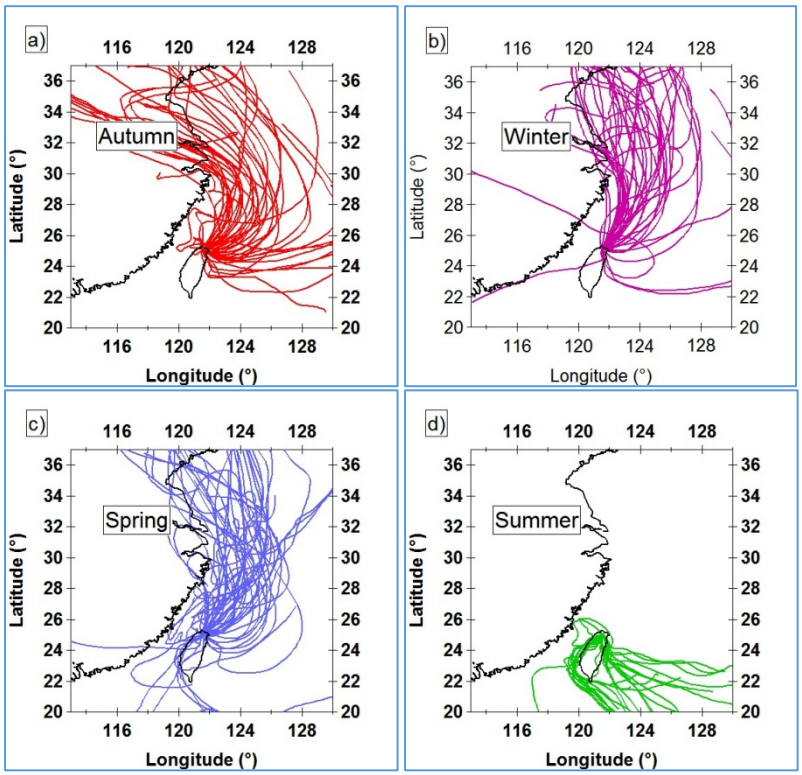
The individual values of the condensation sink, UVB and SO2 have been included in Table 3.

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.

Seasonal variations of ultrafine and sub-micron aerosols in Taipei, Taiwan: implications for particle formation processes in urban areas

Cheung, H. C.[1], Chou, C. C.-K.[1,*], Chen, M.-J.[1], Huang, W.-R.[1], Huang, S.-H.[1], Tsai, C.-Y.[1], Lee, C. S. L.[2]

1. Research Center for Environmental Changes, Academia Sinica, Taipei 11529, Taiwan
2. Institute of Occupational Medicine and Industrial Hygiene, College of Public Health, National Taiwan University, Taipei, Taiwan

**Correspondence to*: C. C.-K. Chou (ckchou@rcec.sinica.edu.tw)

Abstract

The aim of this study is to investigate the seasonal variations in the physicochemical properties of atmospheric ultrafine particles (UFPs, d ≤ 100nm) and submicron particles (PM_1, d ≤ 1μm) in an East-Asian urban area, which are hypothesized to be affected by the interchange of summer and winter monsoons. An observation experiment was conducted at the TARO (Taipei Aerosol and Radiation Observatory), an urban aerosol station in Taipei, Taiwan, from October 2012 to August 2013. The measurements included the mass concentration and chemical composition of UFPs and PM_1, as well as the particle number concentration (PNC) and number size distribution (PSD) with size range of 4-736 nm. The results indicated that the mass concentration of PM_1 was elevated during cold seasons with peak level of 18.5 μg m^{-3} in spring, whereas the highest UFPs concentration was measured in summertime with a mean of 1.64 μg m^{-3}. Moreover, chemical analysis revealed that the UFPs and PM_1 were characterized by distinct composition; UFPs were composed mostly of organics, whereas ammonium and sulfate were the major constituents in PM_1. The seasonal median of total PNCs ranged from 13.9×10^3 cm^{-3} in autumn to 19.4×10^3 cm^{-3} in spring. The PSD information retrieved from the corresponding PNC measurements indicated that the nucleation mode PNC ($N_{4\text{-}25}$) peaked at 11.6×10^3 cm^{-3} in winter, whereas the Aitken mode ($N_{25\text{-}100}$) and accumulation mode ($N_{100\text{-}736}$) exhibited summer maxima at 6.0×10^3 and 3.1×10^3 cm^{-3}, respectively. The change in PSD during summertime was attributed to the enhancement in the photochemical production of condensable organic matter that, in turn, contributed to the growth of aerosol particles in the atmosphere. In addition, clear photochemical production of particles was observed, mostly in summer season, which were characterized by averaged particle growth and formation rates of 4.0±1.1 nm h^{-1} and 1.4±0.8 cm^{-3} s^{-1}, respectively. The prevalence of new particle formation (NPF) in summer was

suggested as a result of seasonally enhanced photochemical oxidation of SO_2 that contributed to the production of H_2SO_4, and low level of PM_{10} ($d \leq 10\mu m$) that served as the condensation sink. Regarding the sources of aerosol particles, correlation analysis upon the PNCs against NO_x revealed that the local vehicular exhaust was the dominant contributor of the UFPs throughout the year. On the contrary, the Asian pollution outbreaks had significant influence in the PNC of accumulation mode particles during the seasons of winter monsoons. 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.

1. *Introduction*

Due to the significant impact of particulate matter on human health and climate change, it is vital to understand the formation process of atmospheric particles (Charlson et al., 1992; Donaldson et al., 1998). A number of mechanisms have been proposed by which atmospheric particles are formed, including binary nucleation, ternary nucleation and ion-induced nucleation for charged particles, under different environment conditions (Kulmala 2003; Kulmala et al., 2004, 2012). Numerous studies have been conducted in different locations to elucidate particle formation processes under various environmental settings in the free troposphere, boreal forest and coastal areas, where new particles formation processes are observed frequently (Kulmala et al., 2004, Holmes 2007). Recently, investigations were also carried out on new particle formation within urban boundary layer (e.g., Cheung et al., 2013 and references therein), where particle formation was suggested to be mainly influenced by the photo-oxidation of SO_2. Furthermore, formation of particulate matter by heterogeneous reactions of gases on dust particles was reported recently (Hsu et al., 2014, Nie et al., 2014). Previous investigations have indicated that the air pollutants, both in gaseous and particulate form, associated with the continental outflows of air masses could have affected a wide region in East Asia and caused severe regional air pollution (e.g., Lin et al., 2004; Wang et al., 2003). However, the formation processes of ultrafine particles (UFPs, $d \leq 100$nm) and sub-micron particles (PM_1, $d \leq 1\mu$m) under the influences of continental outflows are not yet well understood.

In urban environment, major contributing sources of aerosol particles include vehicular exhausts (e.g., Pey et al., 2008; Pérez et al., 2010), industrial emissions (Gao et al., 2009) and new particle formation by photochemical reactions (e.g., Pey et al., 2009). Approximately 55-69% of the total particle number concentrations (PNCs) were attributed to secondary aerosols during midday in several European cities (Reche et al., 2011). In Taipei, Taiwan, a subtropical urban area, Cheung et al. (2013) observed that there was a ten-fold increase in nucleation mode particle number concentrations ($N_{9\text{-}25}$, with size $9 < d < 25$nm) during new particle formation events compared to that contributed by the vehicle emission. Besides the local sources, air quality of East Asian countries is also strongly affected by the transport of air pollutants from mainland China during periods of winter monsoons (Cheung et al., 2005; Lin et al., 2004; Matsumoto et al., 2003). Lin et al. (2004) reported that the mass concentration of particulate matter (PM_{10}) due to the long-range transport associated with winter monsoons was 85 μg m^{-3}, about 79% higher than that due to local pollution (~47.4μg m^{-3}) in urban Taipei. Chemical composition of fine and coarse particles was measured during a winter monsoon period at Rishiri Island, near

the northern tip of Japan, to study the transport of continental aerosols (Matsumoto et al., 2003). The results showed that higher levels of particle mass concentrations were associated with the outbreaks of continental polluted air masses. In addition, Cheung et al. (2005) found deterioration in visibility around the southern China during wintertime as indicated by a two-fold increase in aerosol light scattering coefficient under the influences of winter monsoons. All these studies were limited to measurements in terms of PM_{10} or $PM_{2.5}$ for a particular period, and the seasonal variations of particles in either ultrafine or sub-micron range have not been well illustrated.

A 1-year aerosol characterization experiment was conducted in the 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. In this study, we analyzed number concentration and size distribution of aerosol particles, together with the mass concentration and chemical composition of UFPs and PM_1 measured during four seasonal campaigns (i.e. 24 Oct – 15 Nov 2012, 4 – 24 Jan, 17 Mar – 11 Apr, and 1 – 14 Aug 2013). The results of this study will contribute to the management strategies of the severe air pollution over the East Asia region.

2. Methodology

2.1 Observation site and instrumentation

The measurements were conducted at the Taipei Aerosol and Radiation Observatory (TARO, 25.02 N, 121.53 E), located in the downtown area of Taipei, Taiwan, during October 2012 to August 2013. The measurements were carried out for 2 – 3 weeks in each season (see **Table 1** for measurement details). The aerosol observatory locates on the top floor of the Building-B of the Department of Atmospheric Sciences, National Taiwan University (ASNTU), which is ~20 m a.g.l. (Cheung et al., 2013).

Particle number size distribution (PSD) in the range of 4 – 736 nm was measured by two scanning mobility particle sizer (SMPS) systems. One was equipped with a long-differential mobility analyzer (long-DMA, Model: TSI 3081, TSI Inc.) and a condensation particle counter (CPC) (Model: TSI 3022A, TSI Inc.) to measure the particles from 10 – 736 nm, which was named long-SMPS. Another one was equipped with a nano-DMA (Model: TSI 3085, TSI Inc.) and an ultrafine water-based CPC (UWCPC, Model: TSI 3786, TSI Inc.) to measure the particles from 4 – 110 nm, which was named nano-SMPS. The poly-disperse particles were classified into

selected mono-disperse particles according to their electric mobility by the DMAs. The number concentration of the mono-disperse particles was then counted by the CPCs. Ambient air was drawn into the SMPS systems from outside the building through a 0.635 cm (inner diameter) conductive tube, and a sampling duration of 5 min was adopted for each PSD measurement. The SMPS systems' flow rates were checked weekly during the sampling period and the accuracy of the particle sizing of the DMAs was checked using polystyrene latex (PSL) spheres before the campaigns. Operation details are referred to Cheung et al. (2013).

Size segregated aerosol samples were collected by a pair of Micro-Orifice Uniform Deposition Impactors (MOUDI, Model: 110, MSP Corp.). Taking the advantage that the cut diameter of the 9th MOUDI impaction stage was exactly 100 nm, the 10th impaction stage (cut diameter = 56 nm) of each MOUDI was removed to allow the after filter function as a collector of UFPs (Marple et al., 1991). The sampling flow rate of MOUDI sampler was 30 lpm. 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. For both UFPs and PM_1 sampling arrangements, one of the paired samplers was equipped with Teflon filters, whereas another was equipped with quartz fiber filters. The Teflon filter samples were used for gravimetric measurement. The quartz filter samples were deployed for analysis of soluble ions (Na^+, NH_4^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, NO_2^-,NO_3^-, SO_4^{2-}) using ion chromatograph (IC), and carbonaceous components (i.e. OC and EC) in the aerosols using a DRI-2001A carbonaceous aerosol analyzer with IMPROVE-A protocol (Chow et al., 2007). Details of the in-lab analysis are as described previously (Salvador and Chou, 2014). Both the PM_1 and UFPs were collected with double-layered quartz filters (i.e. QBQ setup) and the artifacts due to adsorption of gaseous components were corrected as suggested by Subramanian et al. (2004). The sampling duration of each sample set (for both MOUDI and PQ-200 samplers) was from 14:00 - 12:00 LT (22 hr), 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).

Moreover, to assist the data interpretation, the hourly averaged mass concentration of PM_{10}, the mixing ratio of trace gases (i.e. NO_x, SO_2 and O_3) and the meteorology parameters (i.e. wind direction/speed and UVB) from the Guting air quality station of Taiwan Environmental Protection Agency, which is about 1 km from the TARO, were analyzed in this study. The details of instrumentation setup for trace

gases measurement are referred to Cheung et al. (2013).

2.2 Data processing and analysis

The PSD of 4 – 736 nm presented in this study was combined from two sets of SMPS data, where the nano-SMPS corresponded to the size range of 4 – 49.6 nm, and the long-SMPS corresponded to the size > 49.6 nm. The diffusion loss of the particles during the sample transport in the tubing was corrected according to the algorithm proposed by Holman (1972). Particle number concentrations for different size ranges were then calculated from the SMPS measurements.

The 5-min PSD data were synchronized into hourly averages, and fitted by the DO-FIT model developed by Hussein et al. (2005) according to the multiple log-normal distribution algorithms. Based on the fitted PSD data, the PNCs were classified into $4 \leq d \leq 25$ nm ($N_{4\text{-}25}$), $25 < d \leq 100$ nm ($N_{25\text{-}100}$), $4 \leq d \leq 100$ nm ($N_{4\text{-}100}$), $100 < d \leq 736$ nm ($N_{100\text{-}736}$) and $4 \leq d \leq 736$ nm ($N_{4\text{-}736}$), for nucleation mode, Aitken mode, ultrafine, accumulation mode and total particles, respectively. Pearson correlation coefficient, *r*, was calculated by *PASW Statistics ver. 18* (SPSS Inc.) to determine the correlation between the respective parameters.

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 or 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 ($dN_{4\text{-}25}/dt$) 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).

2.4 Back-trajectory analysis

Backward trajectories were calculated using the HYSPLIT model (Hybrid Single Particle Lagrangian Integrated Trajectory, Version 4.9) of NOAA (National Oceanic

and Atmospheric Administration) (Draxler, 1999) for TARO during the sampling period, in order to trace the origins of the air masses. 72-h back trajectories were calculated twice per day at 00:00LT and 12:00LT with height setting of 200 m above ground level. It should be noted that the grid resolution of the meteorological data used for back-trajectories calculation is 1° x 1°, which is not enough to trace the detailed air mass passage over the scale of the study region and, therefore, the trajectories only provide an indication of the region from which the air mass was originated.

3. *Results and discussions*

3.1 Particle number concentration and size distribution in respective seasons

As mentioned above, the air quality of urban Taipei is significantly affected by both the local vehicular exhausts and long-range transport of pollution, where the later is dominated by meteorological factors. The information on the meteorological conditions, particularly the wind patterns, is important to elucidate our results and thus presented here. The back-trajectories of the air masses for the TARO are illustrated in **Figure 1 (left panel)**. The results showed that northeasterly winds prevailed in autumn and winter seasons, passing through the Asian continent before reaching Taiwan, whereas southerly winds prevailed in summertime. The air masses observed in spring period were found to be mainly associated with Asian continental outflows and occasionally with the southerly flows. This observation agreed with the surface wind direction measured in urban Taipei area (see **Figure 1, right panel**), where northeasterly winds were dominating during the period from November 2012 to May 2013, and southerly winds were prevailing from May 2013 to August 2013.

The particle number concentrations in various size ranges during each season are summarized in **Table 1**. Relatively higher total PNCs ($N_{4\text{-}736}$) were observed in spring and winter with median values of 19.4×10^3 and 17.4×10^3 cm^{-3}, respectively, followed by that of summer (16.6×10^3 cm^{-3}) and autumn (13.9×10^3 cm^{-3}). This result is comparable to the previous measurements conducted in urban Taipei where the seasonal means of PNCs ($10 < d < 560$nm) ranged from 11.0×10^3 to 17.0×10^3 cm^{-3} (Cheng et al. 2014). **Figure 2** illustrates the number, surface and volume size distributions of the aerosol particles. The geometric mean diameter (GMD) of each PSD mode was retrieved from the data of number concentration. The GMDs of the nucleation, Aitken and accumulation modes were found to be 10.4-12.8 nm, 26.5-38.4 nm, and 91.8-159.0 nm, respectively.

In addition, the fitted GMDs of surface distribution were found to be 77.4 and 293 nm 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 372 nm 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.

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 the 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.

3.2 Mass concentration and chemical composition

Figures 3a and **3b** illustrate the averaged chemical composition and mass concentration of UFPs and PM_1, respectively, for each season. Details of the mass concentration and chemical composition of UFPs and PM_1 are listed in Table S1 in the appendix.

The seasonal means of UFPs ranged from 0.73 to 1.64 μg m^{-3}, with an annual average of 1.01 μg m^{-3}. The measured UFPs mass concentration of the present study was comparable to that in urban area of Los Angeles, United States (0.80 – 1.58 μg m^{-3}, Hughes et al. 1998), and relatively higher than that in urban Helsinki, Finland

(average: 0.49 μg m^{-3}, Pakkanen et al. 2001). 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%). In addition, a large fraction of mass was contributed by the group of "others", which consisted of mineral (K^+, Ca^{2+}, PO_4^{3-} and Mg^{2+}), sea-salt (Na^+ and Cl^-), and unidentified species. The results showed that, on average, mineral and sea salt components attributed only 3.5 % (seasonal means: 2.0-6.0 %) to UFPs mass concentration. Thus a substantial amount of UFPs remained unidentified, which likely included hydrogen and oxygen associated with organic carbon (OC). The conversion factors used to estimate the average molecular weight per carbon in particulate organic matter varied depending on the characteristic of aerosols. A lower factor value, 1.2, was usually suggested for saturated organic molecules, while a higher value, 1.6, was adopted for water-soluble compounds consisting of multifunctional oxygenated groups, and even higher factor values were suggested for aged aerosols which contained higher portion of low and semi-volatile products of photochemical reactions (Turpin and Lim et al. 2001). The high un-identified mass fraction implied that the photochemical production of secondary organic aerosols was a significant process responsible for the elevated UFPs levels observed in this study.

As shown in Fig. 3b, average PM_1 was estimated to be 14.7 μg m^{-3} (seasonal means: 11.6-18.5 μg m^{-3}) in this study, which is similar to the results of a previous study in urban Taipei (average: 14.0 μg m^{-3}, Li et al., 2010). The measured PM_1 level is relatively higher than that of the urban areas of Phoenix, United States (5.9 μg m^{-3}, Lundgren et al. 1996) and Helsinki, Finland (6.1 μg m^{-3}, Vallius et al. 2000). For chemical composition, sulfate was the major mass contributor of PM_1 (average: 39.0 %, seasonal means: 33.8 - 46.8 %), followed by ammonium (average: 12.7 %, seasonal means: 12.0 - 13.2 %) and OC (average: 11.5 %, seasonal means: 9.2 to 14.3 %).

The results presented above indicated that UFPs exhibited a distinct seasonal variability and composition from PM_1 in the study area. The highest UFPs concentration was observed in summer (1.64 μg m^{-3}) and the lowest in winter (0.73 μg m^{-3}). This result may be attributed to the stronger photochemical activities in summer which could have enhanced the formation of secondary organic aerosols. Consequently, the mass concentration of OC increased from 0.20 μg m^{-3} in winter to

0.47 μg m^{-3} in summertime. It is noteworthy that the mass concentration of sulfate in UFPs also peaked in summer (64 ng m^{-3}), suggesting enhancement in photo-oxidation of SO_2. Cheung et al. (2013) found that photo-oxidation of SO_2 was the major mechanism for the formation of new particles in Taipei, Taiwan and the production of condensable vapors was also dominated by photo-oxidation. The co-variations in sulfate and OC revealed in this study further suggested that secondary organic compounds were the major condensable matter contributing to the growth of newly formed particles.

While the organics predominated in the mass concentration of UFPs, which included nucleation mode and Aitken mode particles, the measurements of PM_1 in this study suggested that sulfate was the major constituent of accumulation mode aerosols. In contrast to the seasonal variation of UFPs, the mass concentration of PM_1 reached the maximum at 18.5 μg m^{-3} in spring and exhibited the minimum at 11.6 μg m^{-3} in summer. The PM_1 differences between spring and summer were mostly due to declined ambient levels of sulfate, nitrate, and ammonium ions. As a result, the mass contribution of the three inorganic ions in PM_1 reduced from 55.7 % to 46.2 % and, on the contrary, the mass fraction of OC increased from 10.2 to 14.3 %. The seasonal characteristics of PM_1 concentration and composition were 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**). Our previous studies reported that the fine particulate matter ($PM_{2.5}$) transported on the Asian outflows to northern Taiwan maximized in springtime and were enriched in sulfate, nitrate, and ammonium (Chou et al., 2008; 2010). The seasonal variability of PM_1 found in this study was consistent with the previous observations for $PM_{2.5}$ and thereby suggested the significance of Asian outflow aerosols to the PM_1 budget in the downwind areas of the Asia Continent.

3.3 Seasonal characteristics of photochemical production

In order to study the influences of photochemical production of particles, the measurements of PNC and PSD were analyzed per daytime (07:00 – 17:00 LT) and nighttime (17:00 – 07:00 LT), respectively (see **Figure 4**). In urban environment, the possible sources influencing the PNC and PSD are complicated, which include not only the direct emission from primary sources but also interaction between the newly formed particles, pre-existing particles and condensing vapors through the condensation and coagulation processes. Nevertheless, these processes occurred throughout the day and will not dominate in the differences between daytime and nighttime PNCs as observed in this study. It was assumed that photochemical reaction

was the major attributing factor to the observed diurnal differences in PNC. Since the particles in nighttime were mainly emitted from the vehicular exhausts and the elevated PNCs in daytime were due to both the primary and secondary sources of the particles in the study area (Cheung et al., 2013), a larger difference between the PNCs observed in daytime and nighttime indicated stronger influences of photochemical production on the PNCs. The most striking seasonal features shown in **Figure 4** is the large difference between daytime against nighttime PSD in summer as indicated by the low $N_{4\text{-}736}$ (nighttime)/$N_{4\text{-}736}$ (daytime) ratio, whereas higher ratios were observed 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.

3.4 Factors affecting new particle formation (NPF)

As shown in previous study, the NPF events were frequently observed in summer, which subsequently induced a notable increase in $N_{4\text{-}25}$ 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 $N_{4\text{-}25}$ against NO_x 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 cities 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**.

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 (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**). 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.

The scatter plot between UVB*SO_2 and condensation sink is depicted in **Figure 7**. Relatively higher UVB*SO_2 values were obtained during NPF events. Notably, there was a group of data with high UVB*SO_2/CS but low UVB*SO_2 where no NPF event was observed. This implied that there could be a threshold level of UVB*SO_2 for NPF in the study region. However, some exceptions existed in the dataset and suggested that the parameters driving NPF have not been well accounted and need to be further studied. It was also noticed that an Asian outflow event occurred on 7 April 2013 during which an atypical NPF was observed (labeled as black dot in **Figure 7**). This could be relevant to the secondary particle formation on dust surface under the influence of long-range transport of air mass. This will be discussed in further details in section 3.6.

3.5 Influences of local emission on PNCs

Vehicle emission is known as the major source of the particulate matter in urban environment, particularly during the nighttime. In order to investigate the relationship between the vehicular exhausts and PNCs, the scatter plots of NO_x (as an indicator of vehicle emission) against $N_{4\text{-}25}$, $N_{25\text{-}100}$ and $N_{100\text{-}736}$ during the nighttime were examined for winter and summer periods (see **Figure 8**). The values of the Pearson correlation coefficient (r) and the slope of linear regression between NO_x and PNCs are summarized in **Table 4**.

The highest r values were found in both the plots of NO_x against $N_{25\text{-}100}$ for winter (r = 0.88) and summer (r = 0.87). This result suggested a strong linear correlation between the vehicle emission and the $N_{25\text{-}100}$ which coincided with the

results from previous studies (e.g., Morawska et al. 2008). During wintertime, stronger correlation was found between NO_x against $N_{4\text{-}25}$ (r = 0.84) and $N_{25\text{-}100}$ (r = 0.88) compared to that between NO_x and $N_{100\text{-}736}$ (r = 0.38). In contrast, high r values were obtained between NO_x and all particle modes in summer (r = 0.70 - 0.87). The robust correlation of NO_x and $N_{4\text{-}25}$, also NO_x and $N_{25\text{-}100}$ suggested that local vehicle emission was the predominant source of UFPs throughout a year. These results coincided with previous studies on the size distribution of vehicle exhaust particles, which were found to be 20-130 nm and 20-60 nm, respectively, for diesel and petrol engine vehicles (Harris and Maricq 2001, Ristovski et al. 2006). However, the PNCs of accumulation mode particles ($N_{100\text{-}736}$) in winter were dominated by NO_x-independent sources, which were most likely related to the pollution outbreaks from the Asian continent. Lin et al. (2004) indicated that the long-range transported air mass was characterized by high level of PM_{10} and low mixing ratio of NO_x due to its short atmospheric lifetime. Interestingly, moderate correlation between the PNC of accumulation mode particles ($N_{100\text{-}736}$) and NO_x was also observed in summer. Given that the Asian outflow was ceased in summertime, this correlation evidenced substantial contribution of local sources, particularly vehicular emissions, to the PNC of accumulation mode particles in Taipei, Taiwan.

The slope values can serve as a relative emission factor of particles per NO_x, 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. The seasonal effects on the emission ratio of PNCs and NO_x are rather difficult to address due to the complexity of different controlling factors, such as formation mechanisms and meteorological conditions. For example, Nam et al. (2010) reported negatively exponential correlation between the PM/NO_x ratio in vehicle emission and ambient temperature, and suggested that the impact of ambient temperature on particulate matter was larger than that on NO_x. Nevertheless, the observed differences in the $PNCs/NO_x$ ratios for winter and summer periods of this study necessitate further investigations on the formation mechanisms of aerosol particles in urban areas, in particular the nucleation and the Aitken modes.

3.6 Influence of long-range transport (LRT)

During the seasons of winter monsoons, i.e. from autumn to spring, the continental outflows have been frequently observed in urban Taipei, which is

indicated by the stable northeasterly wind and increase of O_3 level (Lin et al. 2004). Previous studies of long-range transport (LRT) of air pollutants on air quality of northern Taiwan showed that an elevated PM_{10} was observed under the influence of continental outflows (Lin et al., 2004, Chou et al., 2004). **Figure 9** depicts an LRT pollution event observed at the TARO during this study. The wind direction changed from westerly/northwesterly to northeasterly at 21:00, 24 March which continued until 06:00, 26 March. During this period, the O_3 mixing ratio remained at moderate level (~30-55 ppb) and PM_{10} increased from 10.0 to 98.0 μg m^{-3}. 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. In this section, we attempt to analyze the PSD/PNC under the influences of continental pollution outbreaks. The periods of the respective LRT events are listed in **Table S2**.

As shown in **Figure 9**, the diurnal variations of PSD during the LRT event exhibited two $N_{4\text{-}25}$ peaks associated to the morning and afternoon traffic rush hours, whereas the PNCs of the Aitken mode particles remained at a low level. The results 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 observed 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 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).

The averaged PSDs for LRT and non-LRT cases are shown in **Figure 10**. The geometric mean diameters of the nucleation, Aitken, and accumulation modes in PSD were found to be 10.6, 37.2 and 156.8 nm for LRT and 11.3, 30.0 and 113.4 nm for non-LRT cases, respectively. The median $N_{4\text{-}25}$ ($11.1\text{x}10^3$ cm^{-3}), $N_{25\text{-}100}$ ($7.3\text{x}10^3$ cm^{-3}) and $N_{100\text{-}736}$ ($1.8\text{x}10^3$ cm^{-3}) observed in non-LRT events were significantly higher than those for LRT events ($N_{4\text{-}25}$: $9.2\text{x}10^3$ cm^{-3}, $N_{25\text{-}100}$: $3.8\text{x}10^3$ cm^{-3}, $N_{100\text{-}736}$: $1.3\text{x}10^3$ cm^{-3}).

This was attributed to the lower average wind speed (and hence poor dispersion) during non-LRT events (1.5±0.8 m s^{-1}) than that for LRT events (3.0±0.8 m s^{-1}). In contrast to the increase in PM_{10} observed usually during LRT episodes (e.g., Lin et al., 2012), the relatively lower PNCs suggested that the number concentration of submicron particles, in particular UFPs, was dominated by local emissions during the episodes of continental pollution outbreaks. This agreed with the observation of seasonal UFPs mass concentration that peaked in summertime when Taiwan was isolated from the influences of continental air mass.

4. Conclusions

The mass concentration and chemical composition of ultrafine particles (UFPs) and submicron particles (i.e. PM_1) as well as the particle number concentration (PNC) and size distributions (PSD) 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. Significant seasonal variability and chemical composition of UFPs and PM_1 were revealed. The UFPs were composed mostly of organic matter and reached maxima in summer, whereas the PM_1 composition was dominated by ammonium and sulfate and exhibited a seasonal peak in spring.

It was found that the total PNC was significantly elevated during cold seasons, which was caused mostly by the high level of nucleation mode particles ($N_{4\text{-}25}$). On the contrary, both the Aitken mode ($N_{25\text{-}100}$) and accumulation mode ($N_{100\text{-}736}$) PNCs reached their respective maxima in summertime. Consistent correlation without significant seasonal variations was found between the UFPs (i.e. nucleation and Aitken mode particles) and NO_x, suggesting that local vehicle emission was the major source of UFPs in the study area throughout a year. The local vehicle emission was also dominating the accumulation mode PNC in summer, but not in wintertime. The declined correlation between NO_x and $N_{100\text{-}736}$ in winter (r = 0.38) was likely due to the influences of air pollution associated with the Asian outflows.

The elevated UFPs level in summer was attributed to the increase in the concentration of Aitken mode particles ($N_{25\text{-}100}$). 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. Moreover, the chemical measurements suggested that the constituents of the condensed materials in UFPs

were mostly organic matter, implying the significance of secondary organic aerosols in the ambient UFPs.

A total of 10 new particle formation (NPF) events occurred out of 84 measurement days in this study, which were observed in autumn (1 out 23 days), spring (3 out of 26 days) and summer (6 out of 14 days) seasons. The prevalence of NPF in summer agreed with the highest H_2SO_4 proxy and lowest PM_{10} observed in this study, which provided favorable atmospheric conditions for new particle formation. The averaged particle growth and formation rates for the NPF events were 4.0±1.1 nm h^{-1} and 1.4±0.8 cm^{-3} s^{-1}, respectively, which were comparable to those measured in previous urban studies.

As exemplifying above, the characteristics of various physicochemical properties of particles investigated in this study and the occurrence of NPF exhibited a strong seasonal variability, which was co-influenced by the long-range transported particles during the seasons of winter monsoons and the strong photochemical activities in summer. The results of this study are critical for the authorities involved in urban development and health impact assessment, and the environmental policy makers who are tackling the severe atmospheric pollution in the East Asia region.

Acknowledgements

This research was supported by the Academia Sinica and the Ministry of Science and Technology of Taiwan through grants 103-2111-M-001-003, 102-2628-M-001-007, and 101-2119-M-001-003. The authors thank the Taiwan EPA for providing the air quality and meteorological data. We also thank the Department of Atmospheric Science of National Taiwan University for the logistical supports to the operation of TARO. Prof. Tareq Hussein is gratefully acknowledged for providing us the code of DO-FIT.

573 **Table 1.** Median and standard deviation of the PNCs measured in each season. The size
574 ranges of the PNCs were represented by the subscripted number. For example, $N_{4\text{-}25}$,
575 represents the number concentrations of the particles from 4 to 25 nm. The fractions of $N_{4\text{-}25}$
576 and $N_{4\text{-}100}$ to total PNCs were presented in the last two columns.
577

	Measurement periods	$N_{4\text{-}736}$ ***(#/cm^3)***	$N_{4\text{-}25}$ ***(#/cm^3)***	$N_{25\text{-}100}$ ***(#/cm^3)***	$N_{4\text{-}100}$ ***(#/cm^3)***	$N_{100\text{-}736}$ ***(#/cm^3)***	$N_{4\text{-}25}/N_{4\text{-}736}$	$N_{4\text{-}100}/N_{4\text{-}736}$
Autumn	24 Oct–15 Nov2012	13.9 x 10^3 (6.7x10^3)	8.6 x 10^3 (4.5x10^3)	3.9 x 10^3 (2.7x10^3)	12.7 x 10^3 (6.4x10^3)	1.3 x 10^3 (0.9x10^3)	0.62	0.90
Winter	4–24 Jan 2013	17.4 x 10^3 (8.7x10^3)	11.6 x 10^3 (6.0x10^3)	4.1 x 10^3 (3.7x10^3)	16.3 x 10^3 (8.5x10^3)	0.9 x 10^3 (0.9x10^3)	0.70	0.94
Spring	17 Mar–11 Apr 2013	19.4 x 10^3 (13.8x10^3)	10.3 x 10^3 (11.3x10^3)	5.8 x 10^3 (4.3x10^3)	17.0 x 10^3 (13.6x10^3)	1.9 x 10^3 (1.1x10^3)	0.56	0.89
Summer	1–14 Aug 2013	16.6 x 10^3 (16.4x10^3)	6.9 x 10^3 (9.1x10^3)	6.0 x 10^3 (9.1x10^3)	13.7 x 10^3 (15.4x10^3)	3.1 x 10^3 (2.6x10^3)	0.44	0.87

578

579 **Table 2.** Time periods defined as the new particle formation events and the particle growth
580 and formation rates

Date	*Time period (LT)*	*Growth rate (nm h^{-1})*	*Formation rate (cm^{-3} s^{-1})*
9 Nov 2012	07:00-13:00	3.4	1.30
26 Mar 2013	06:00-10:00	3.4	1.91
4 Apr 2013	07:00 – 10:00	3.7	1.13
5 Apr 2013	08:00 – 12:00	5.5	1.10
4 Aug 2013	09:00 – 12:00	3.9	1.84
5 Aug 2013	09:00 – 13:00	4.9	2.44
7 Aug 2013	06:00 – 12:00	3.5	0.84
8 Aug 2013	09:00 – 12:00	5.0	2.76
9 Aug 2013	06:00 – 13:00	1.6	0.39
11 Aug 2013	06:00 – 09:00	4.8	0.58
Average		4.0 (±1.1)	1.4 (±0.8)

581

582

583 **Table 3**. Average of N_{4-25}, PM_{10}, UVB, SO_2, condensation sink (CS), H_2SO_4 proxy and wind
584 speed of different seasons. (note: the data with observation of rainfall was not used in
585 calculation).

Periods	***N_{4-25} (cm^{-3})***	***PM_{10} ($\mu g\ m^{-3}$)***	***UVB (Wm^{-2})***	***SO2 (ppb)***	***CS ($10^{-2}\ s^{-1}$)***	***H_2SO_4 proxy (ppb Wm^{-2} s)***	***Wind speed (ms^{-1})***
Autumn	8.6×10^3	53.9	1.04	2.27	0.85	307.1	2.82
Winter	11.6×10^3	48.4	0.80	2.58	0.75	240.0	2.34
Spring	10.5×10^3	61.1	0.99	2.76	1.35	238.4	2.17
Summer	6.9×10^3	35.6	1.97	3.19	1.89	493.1	2.35

586

587 **Table 4**. Pearson correlation coefficient (*r*) and slope of linear regression of PNCs against
588 NO_x during the nighttime (20:00-04:00 LT) in winter and summer periods.

Periods		$N_{4\text{-}25}$	$N_{25\text{-}100}$	$N_{100\text{-}736}$
Winter	*Slope*	279	163	18
	r	0.84	0.88	0.38
Summer	*Slope*	239	330	155
	r	0.76	0.87	0.70

589

590 **References**

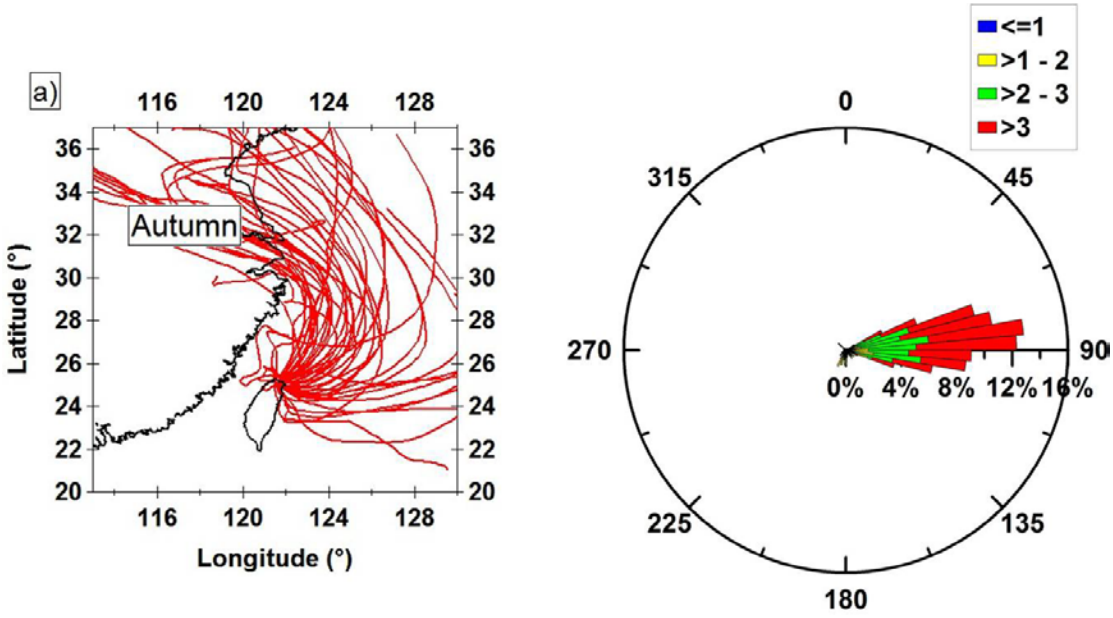
591 Charlson, R.J., Schwartz, S.E., Hales, J.M., Cess, R.D., Coakley Jr., J.A., Hansen, J.E.
592 and Hofmann, D.J. (1992). Climate forcing by anthropogenic aerosols. Science, 255,
593 423-430.
594 Cheng, Y.-H., Kao, Y.-Y. and Liu, J.-J. (2014). Correlations between black carbon
595 mass and size-resolved particle number concentrations in the Taipei urban area: A
596 five-year long-term observation. Atmospheric Pollution Research, 5, 62-72.
597 Cheung, H.C., Wang, T., Baumann, K. and Guo, H. (2005). Influence of regional
598 pollution outflow on the concentrations of fine particulate matter and visibility in the
599 coastal area of southern China. Atmospheric Environment, 39, 6463-6474.
600 Cheung, H.C., Morawska, L. and Ristovski, Z.D. (2011). Observation of new particle
601 formation in subtropical urban environment. Atmospheric Chemistry and Physics, 11,
602 3823-3833.
603 Cheung, H.C., Chou, C.C.-K., Huang, W.-R. and Tsai, C.-Y. (2013). Characterization
604 of ultrafine particle number concentration and new particle formation in an urban
605 environment of Taipei, Taiwan. Atmospheric Chemistry and Physics, 13, 8935-8946.
606 Chou, C. C.-K., Lee, C. T., Cheng, M. T., Yuan, C. S., Chen, S. J., Wu, Y. L., Hsu, W.
607 C., Lung, S. C., Hsu, S. C., Lin, C. Y., Liu, S. C. (2010). Seasonal variations and
608 spatial distribution of carbonaceous aerosols in Taiwan, Atmos. Chem. Phys, 10,
609 9563–9578.
610 Chou, C. C.-K., Lee, C.-T., Yuan, C. S., Hsu, W. C., Hsu, S. C., Liu, S. C. (2008).
611 Implications of the chemical transformation of Asian outflow aerosols for the
612 long-range transport of inorganic nitrogen species. Atmospheric Environment, 42,
613 7508-7519.
614 Chou, C. C.-K., Huang, S.-H., Chen, T.-K., Lin, C.-Y. and Wang, L.-C. (2005).
615 Size-segregated characterization of atmospheric aerosols in Taipei during Asian
616 outflow episodes. Atmospheric Research, 75, 89-109.
617 Chou, C. C.-K., Lin, C.-Y., Chen, T.-K., Hsu, S.-C., Lung, S.-C., Liu, S.C. and Young,
618 C.-Y. (2004). Influence of Long-Range Transport Dust Particles on Local Air
619 Quality: A Case Study on Asian Dust Episodes in Taipei during the Spring of 2002.
620 Terrestrial, Atmospheric and Oceanic Sciences, 15, 881-889.
621 Chow, J. C., Watson, J. G., Chen, L.-W. A., Chang, L.C.O., Robinson, N. F., Trimble,
622 D. and Kohl, S. (2007). The IMPROVE_A Temperature Protocol for
623 Thermal/Optical Carbon Analysis: Maintaining Consistency with a Long-Term
624 Database, Journal of the Air & Waste Management Association, 57:9, 1014-1023.
625 Dal Maso, M., Kulmala, M., Riipinen, I., Wagner, R., Hussein, T., Aalto, P.P. and
626 Lehtinen, K.E.J. (2005). Formation and growth of fresh atmospheric aerosols: eight
627 years of aerosol size distribution data from SMEAR II, Hyytiälä, Finland. Boreal

628 Environ. Res., 10, 323-336.
629 Donaldson, K., Li, X.Y. and MacNee, W (1998). Ultrafine (nanometre) particle
630 mediated lung injury. Journal of Aerosol Science, 29, 553, 560.
631 Draxler, R.R. (1999). HYSPLIT4 user's guide. NOAA Tech. Memo. ERL ARL-230,
632 NOAA Air Resources Laboratory, Silver Spring, MD.
633 Gao, J., Wang, T., Zhou, X. Wu, W. and Wang, W. (2009). Measurement of aerosol
634 number size distributions in the Yangtze River delta in China: Formation and growth
635 of particles under polluted conditions. Atmos. Environ., 43, 829-836.
636 Guo, H., Ding, A., Morawska, L., He, C., Ayoko, G., Li,Y. and Hung, W. (2008). Size
637 distribution and new particle formation in sub-tropical eastern Australia.
638 Environmental Chemistry, 5, 382-390.
639 Harris, S.J. and Maricq, M.M. (2001). Signature size distributions for diesel and
640 gasoline engine exhaust particulate matter. Journal of Aerosol Science, 32, 749-764.
641 Holman, J.P. (1972). Heat Transfer. New York, McGraw-Hill.
642 Holmes, N.S. (2007). A review of particle formation events and growth in the
643 atmosphere in the various environments and discussion of mechanistic implications.
644 Atmospheric Environment, 41, 2183-2201.
645 Hsu, S.-C., Lee, C.S.L., Huh, C.-A., Shaheen, R., Lin, F.-J. and Liu, S.C. (2014).
646 Ammonium Deficiency Caused by Heterogeneous Reactions during a Super Asian
647 Dust Episode. Journal of Geophysical Research, 119(11),
648 doi:10.1002/2013JD021096.
649 Hughes, L.S., Cass, G.R., Gone, J., Ames, M. and Olmez, I. (1998). Physical and
650 Chemical Characterization of Atmospheric Ultrafine Particles in the Los Angeles
651 Area. Environmental Science and Technology, 32, 1153-1161.
652 Hussein, T., Dal Maso, M., Petäjä, T., Koponen, I.K., Paatero, P., Aalto, P.P., Hämeri,
653 K. and Kulmala, M. (2005). Evaluation of an automatic algorithm for fitting the
654 particle number size distributions. Boreal Env. Res. 10:337-355.
655 Juwono, A., Johnson, G.R., Mazaheri, M. and Morawska, L., Roux, F. and Kitchen, B.
656 (2013). Investigation of the airborne submicrometer particles emitted by dredging
657 vessels using a plume capture method. Atmospheric Environment, 73, 112-123.
658 Kulmala, M. (2003). How Particles Nucleate and Grow. Science, 302, 1000-1001.
659 Kulmala, M., Kontkanen, J., Junninen, H., Lehtipalo, K., Manninen, H.E., Nieminen,
660 T., Petäjä, T., Sipilä, M., Schobesberger, S., Rantala, P., Franchin, A., Jokinen, T.,
661 Järvinen, E., Äijälä, M., Kangasluoma, J., Hakala, J., Aalto, P.P., Paasonen, P.,
662 Mikkilä, J., Vanhanen, J., Aalto, J., Hakola, H., Makkonen, U., Ruuskanen, T.,
663 Mauldin III, R.L., Duplissy, J., Vehkamäki, H., Bäck, J., Kortelainen, A., Riipinen I.,
664 Kurtén, T., Johnston, M.V., Smith, J.N., Ehn, M., Mentel, T.F., Lehtinen, K.E.J.,
665 Laaksonen, A., Kerminen, V.-M. and Worsnop, D.R. (2013). Direct Observations of

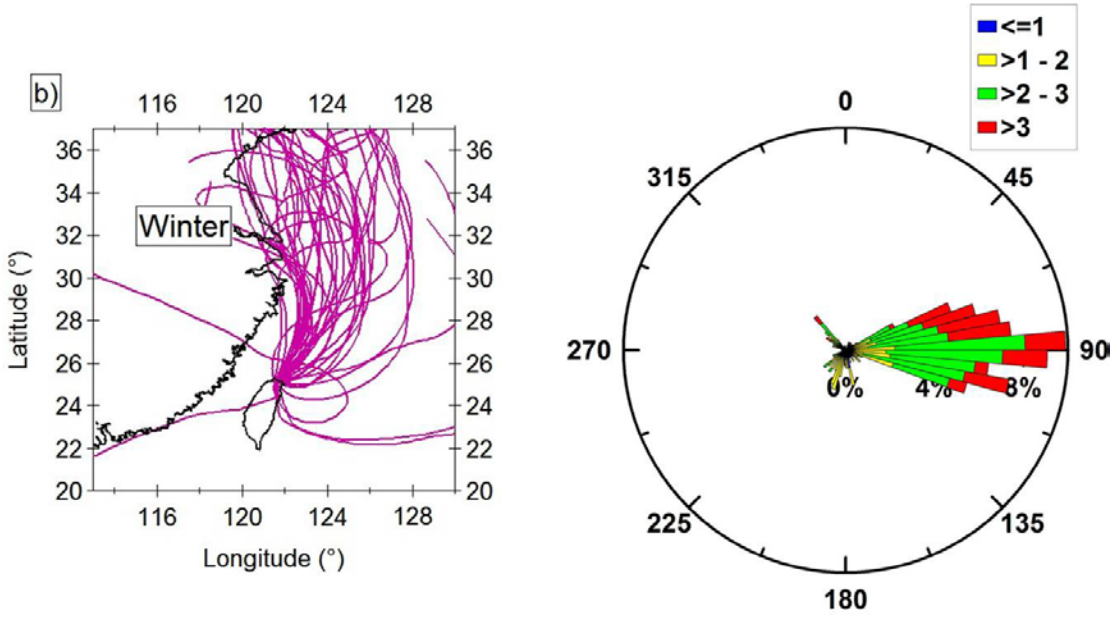
666 Atmospheric Aerosol Nucleation. Science, 339, 943-946.
667 Kulmala, M., Petäjä, T., Nieminen, T., Sipilä, M., Manninen, H.E., Lehtipalo, K., Dal
668 Maso, M., Aalto, P.P., Junninen, H., Paasonen, P., Riipinen, I., Lehtinen, K.E.,
669 Laaksonen, A. and Kerminen, V.-M. (2012). Measurement of the nucleation of
670 atmospheric aerosol particles. Nature Protocols, 7, 1651-1667.
671 Kulmala, M., Vehkamäki, H., Petäjä, T., Dal Maso, M., Lauri, A., Kerminen, V.-M.,
672 Birmili, W. and McMurry, P.H. (2004). Formation and growth rates of ultrafine
673 atmospheric particles: a review of observations. Journal of Aerosol Science, 35,
674 143-176.
675 Lehtinen, K.E.J., Dal Maso, M., Kulmala, M. and Kerminen, V.-M. (2007).
676 Estimating nucleation rates from apparent particle formation rates and vice versa:
677 Revised formulation of the Kerminen-Kulmala equation. J. Aerosol Sci., 38,
678 988-994, doi:10.1016/j.jaerosci.2007.06.009.
679 Li, C.-S. and Lin, C.-H. (2010). PM1/PM2.5/PM10 Characteristics in the Urban
680 Atmosphere of Taipei. Aerosol Science and Technology, 36:4, 469-473.
681 Lin, C.-Y., Liu, S.C., Chou, C.C.-K., Liu, T.H. and Lee, C.-T. (2004). Long-Range
682 Transport of Asian Dust and Air Pollutants to Taiwan. Terr. Atmos. and Ocean. Sci.,
683 15, 759-784.
684 Lin, C.-Y., Chou, C. C.-K., Wang, Z., Lung, S.-C., Lee, C.-T., Yuan, C.-S., Chen,
685 W.-N., Chang, S.-Y., Hsu, S.-C., Chen, W.-C. and Liu, S. C. (2012). Impact of
686 different transport mechanisms of Asian dust and anthropogenic pollutants to Taiwan.
687 Atmos. Environ., 60, 403-418.
688 Lundgren, D.A. Hlaing, D.N., Rich, T.A. and Marlpe, V.A. (1996). $PM_{10}/PM_{2.5}/PM_1$
689 Data from a Trichotomous Sampler. Aerosol Science and Technology, 25, 353-357.
690 Marple , V. A., Rubow, K. L. and Behm, S. M. (1991). A Microorifice Uniform
691 Deposit Impactor (MOUDI): Description, Calibration, and Use, Aerosol Science and
692 Technology, 14:4, 434-446.
693 Matsumoto, K., Uyama, Y., Hayano, T., Tanimoto, H., Uno, Itsushi. and Uematsu, M.
694 (2003). Chemical properties and outflow patterns of anthropogenic and dust
695 particles on Rishiri Island during the Asian Pacific Regional Aerosol
696 Characterization Experiment (ACE-Asia). Journal of Geophysical Research, 108
697 (D23), 8666, doi:10.1029/2003JD003426.
698 Mazaheri, M., Johnson, G.R. and Morawska, L. (2009). Particle and Gaseous
699 Emissions from Commercial Aircraft at Each Stage of the Landing and Takeoff
700 Cycle. Environmental Science and Technology, 43, 441-446.
701 Morawska, L., Ristovski, Z., Jayaratne, E.R., Keogh, D.U. and Ling, X. (2008).
702 Ambient nano and ultrafine particles from motor vehicle emissions: Characteristics,
703 ambient processing and implications on human exposure. Atmospheric Environment,

42, 8113-8138.
Nam, E., Kishan, S., Baldauf, R.W., Fulper, C.R., Sabisch, M. and Warila, J. (2010). Temperature Effects on Particulate Matter Emissions from Light-Duty, Gasonline-Powered Motor Vehicles. Environmental Science and Technology, 44, 4672-4677.
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.
Pakkanen, T.A., Kerminen, V.-M., Korhonen, C.H., Hillamo, R.E., Aarnio, P., Koskentalo, T. and Maenhaut, W. (2001). Urban and rural ultrafine ($PM_{0.1}$) particles in the Helsinki area. Atmospheric Environment, 35, 4593-4607.
Pérez, N., Pey, J., Cusack, M., Reche, C., Querol, X., Alastuey, A. and Viana, M (2010). Variability of Particle Number, Black Carbon, and PM10, PM2.5 and PM1 Levels and Speciation: Influence of Road Traffic Emissions on Urban Air Quality. Aerosol Science and Technology, 44, 487-499.
Pey, J., Rodíguez, S., Querol, X., Alastuey, A., Moreno, T., Putaud, J.P., and Van Dingenen, R (2008). Variations of urban aerosols in the western Mediterranean. Atmospheric Environment, 42, 9052-9062.
Pey, J., Querol, X., Alastuey, A., Rodiguez, S., Putaud, J.P. and Van Dingenen, R. (2009). Source apportionment of urban fine and ultrafine particle number concentration in a Western Mediterranean city. Atmospheric Environment, 43, 4407-4415.
Reche, C., Querol, X., Alastuey, A., Viana, M., Pey, J., Moreno, T., Rodríguez, S., González, Y., Fernández-Camacho, R., de la Rosa, J., Dall'Osto, M., Prévôt, A.S.H., Hueglin, C., Harrison, R.M. and Quincey, P. (2011). New considerations for PM, Black Carbon and particle number concentration for air quality monitoring across different European cities. Atmospheric Chemistry and Physics, 11, 6207-6227, 2011.
Ristovski, Z.D., Jayaratne, E.R., Lim, M., Ayoko, G.A. and Morawska, L. (2006). Influence of diesel fuel sulphur on the nanoparticle emissions from city buses. Environmental Science and Technology, 40, 1314-1320.
Salvador, C.M. and Chou, C.C.-K. (2014). Analysis of semi-volatile materials (SVM) in fine particulate matter. Atmospheric Environment, 95, 288-295.
Subramanian, R., Khlystov, A.Y., Cabada, J.C. and Robinson, A.L. (2004). Positive and negative artifacts in particulate organic carbon measurements with denuded and undenuded sampler configurations. Aerosol Science and Technology, 38, 27-48.
Turpin, B.J. and Lim, H.-J. (2001). Species contributions to PM2.5 mass concentrations: revisiting common assumptions for estimating organic mass. Aerosol

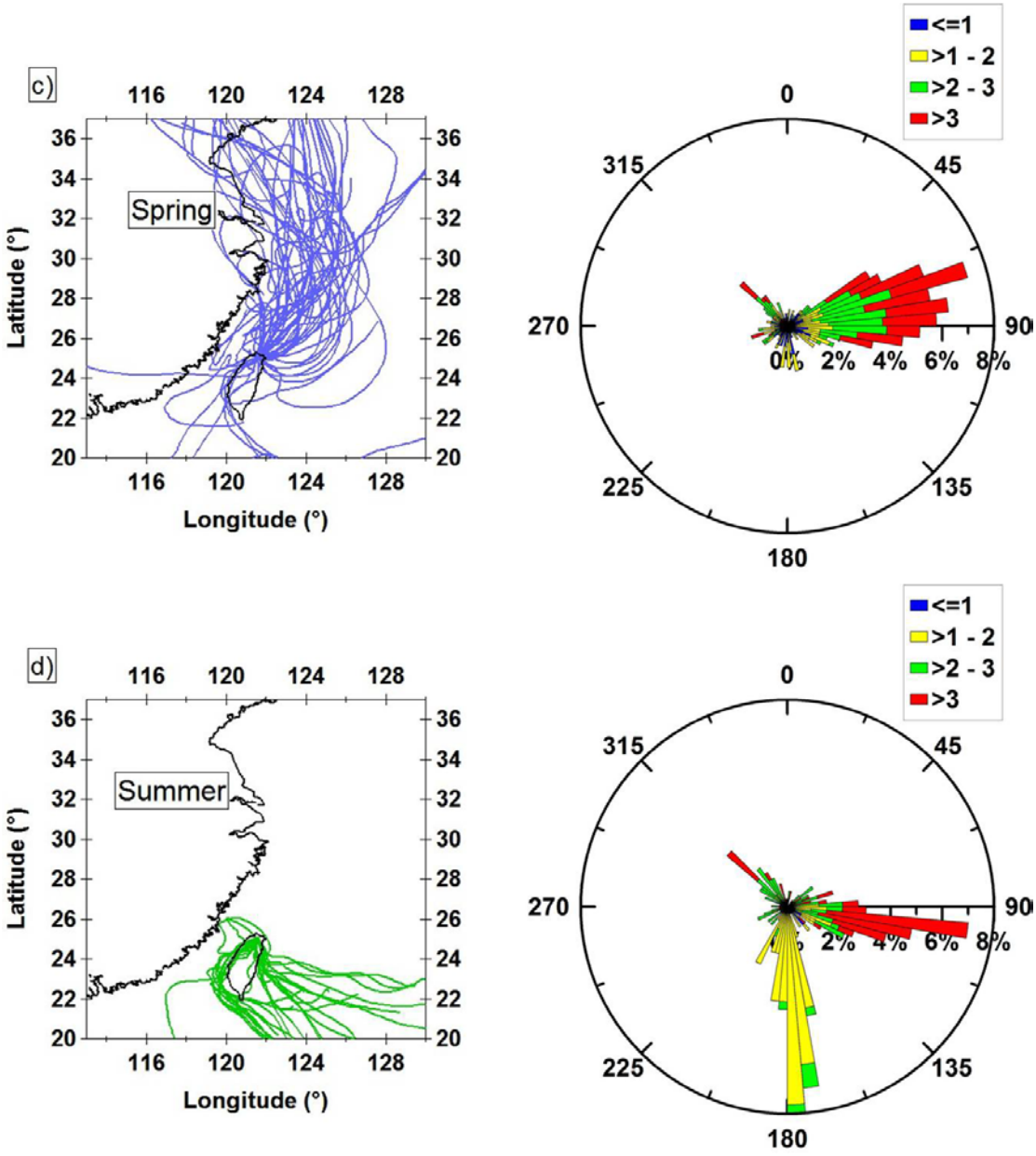
Science and Technology, 35, 602-610.
Vallius, M.J. Ruuskanen, J. Mirme, A. and Pekkanen, J. (2000). Concentrations and Estimated Soot Content of PM_1, $PM_{2.5}$ and PM_{10} in a Subarctic Urban Atmosphere. Environmental Science and Technology, 34, 1919-1925.
Wang, T., Ding, A.J., Blake, D.R., Zahorowski, W., Poon, C.N. and Li, Y-S. (2003). Chemical characterization of the boundary layer outflow of air pollution to Hong Kong during February-April 2001.
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.
Woo, K.S., Chen, D.R., Pui, D.Y.H. and McMurry, P.H. (2001). Measurement of Atlanta aerosol size distribution: observation of ultrafine particle events. Aerosol Science and Technology, 34, 75-87.


a)
Autumn
Latitude (°)
Longitude (°)
116
120
124
128
20
22
24
26
28
30
32
34
36
<=1
>1 - 2
>2 - 3
>3
0
45
90
135
180
225
270
315
0%
4%
8%
12%
16%

760


b)
Winter
Latitude (°)
Longitude (°)
116
120
124
128
20
22
24
26
28
30
32
34
36
<=1
>1 - 2
>2 - 3
>3
0
45
90
135
180
225
270
315
0%
4%
8%

761



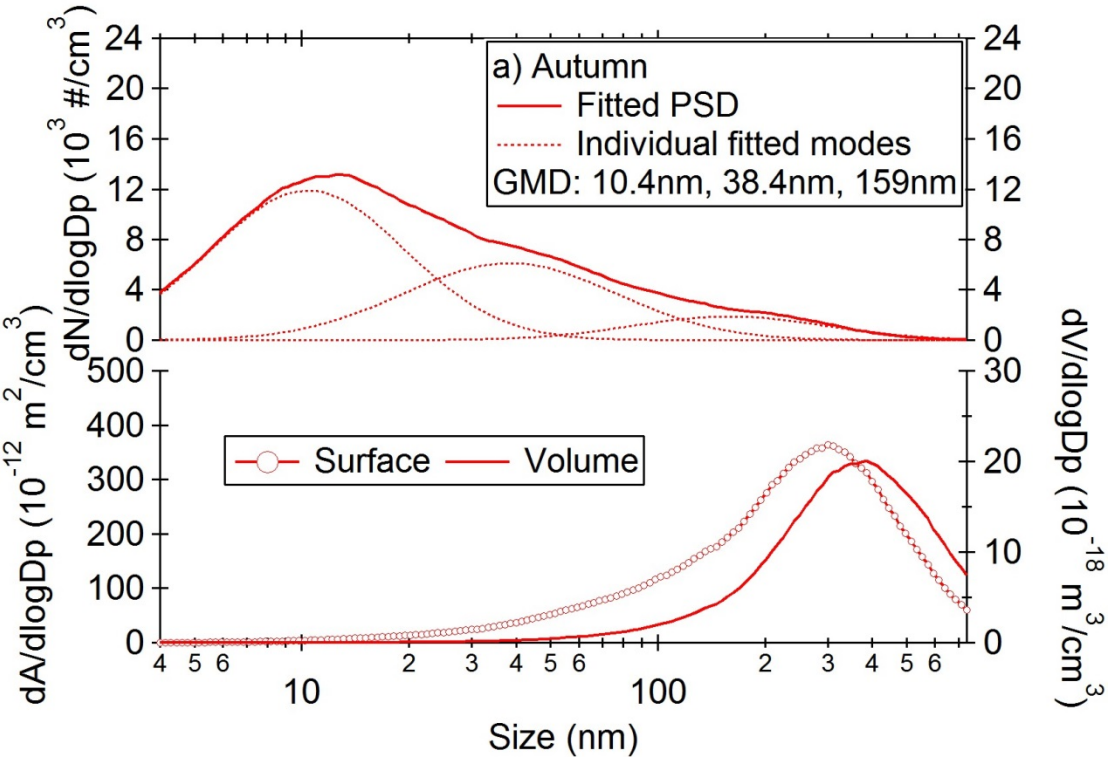
762

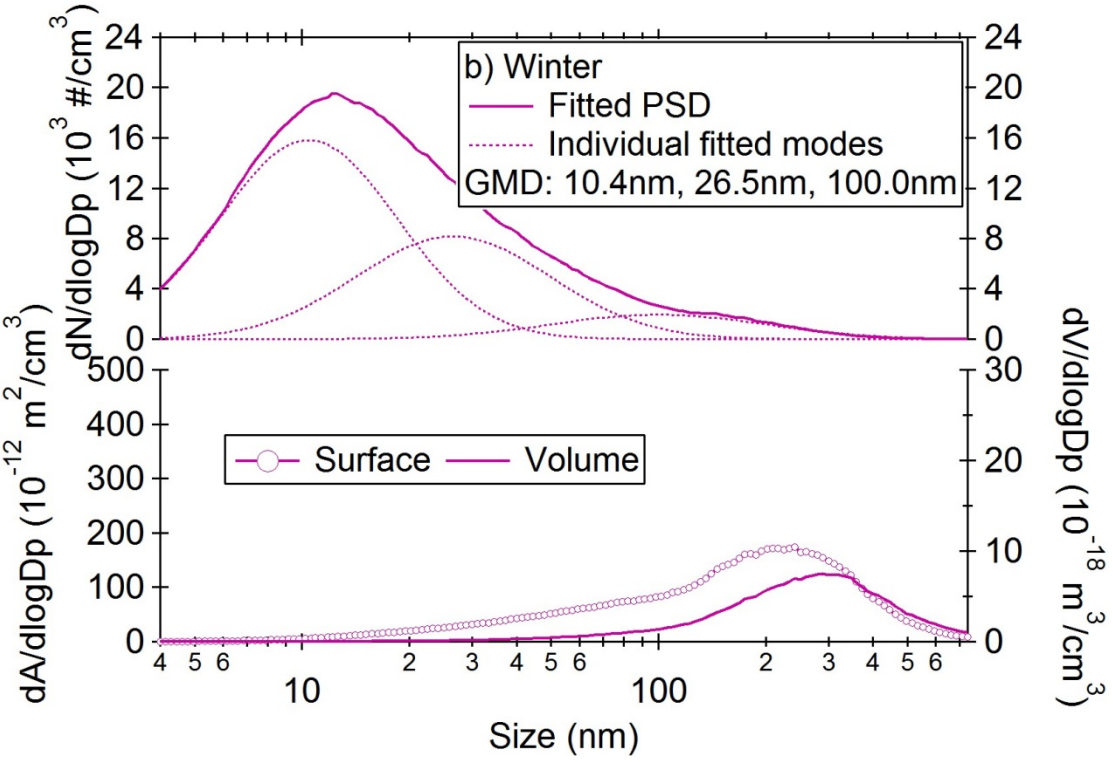
763

764

765 **Figure 1**. Back-trajectories calculated for TARO for the measurement periods (left panel) and
766 surface wind rose plots (right panel) in **(a)** autumn, **(b)** winter, **(c)** spring and **(d)** summer. The
767 color codes of wind rose plots represent the wind speed: blue < 1 ms^{-1}; yellow 1-2 ms^{-1}; green
768 2-3 ms^{-1}; and red > 3 ms^{-1}.

769


a) Autumn
Fitted PSD
Individual fitted modes
GMD: 10.4nm, 38.4nm, 159nm
dN/dlogDp (10^3 #/cm^3)
dA/dlogDp (10^-12 m^2/cm^3)
dV/dlogDp (10^-18 m^3/cm^3)
Surface
Volume
Size (nm)


b) Winter
Fitted PSD
Individual fitted modes
GMD: 10.4nm, 26.5nm, 100.0nm
dN/dlogDp (10^3 #/cm^3)
dA/dlogDp (10^-12 m^2/cm^3)
dV/dlogDp (10^-18 m^3/cm^3)
Surface
Volume
Size (nm)

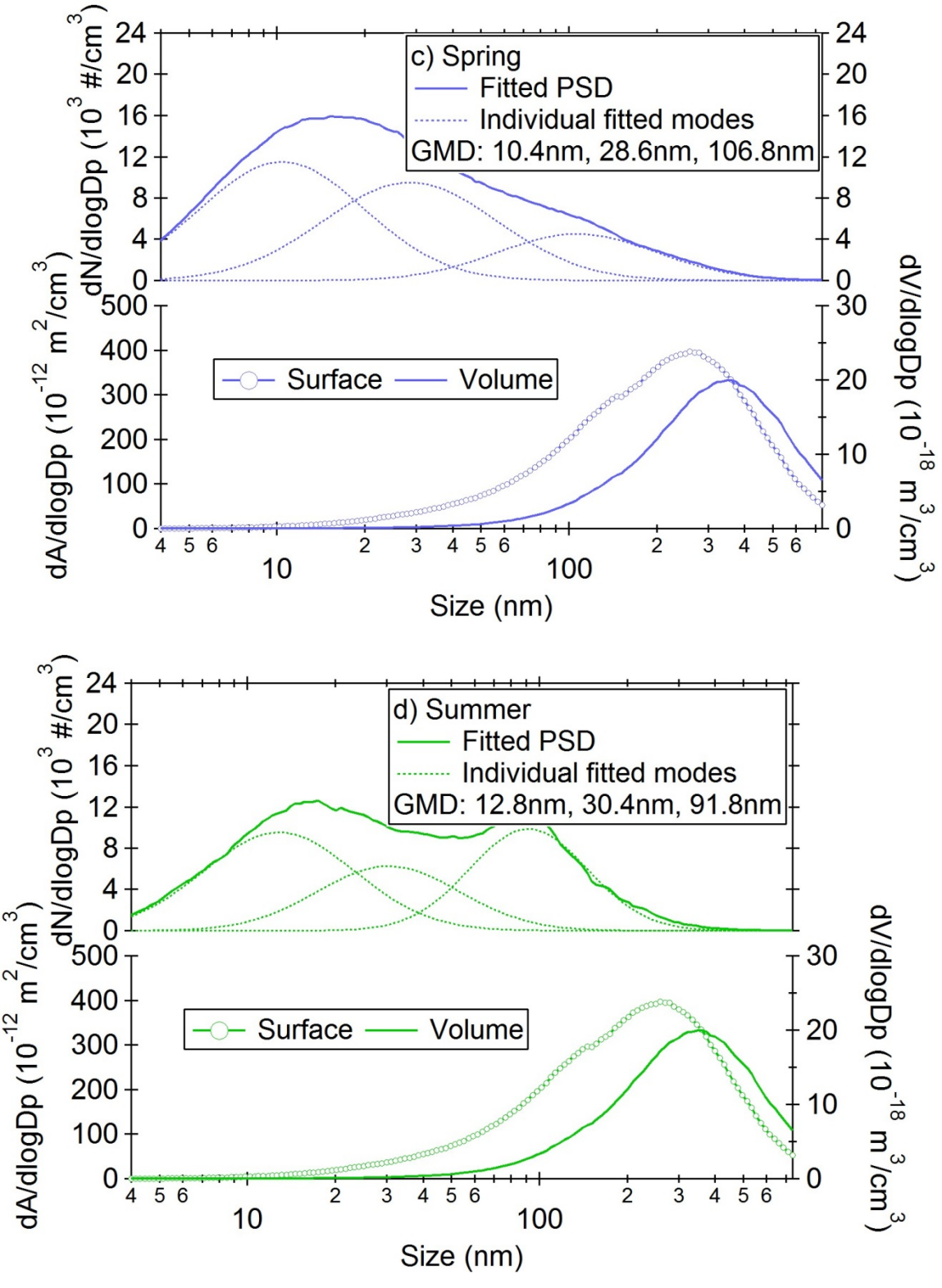
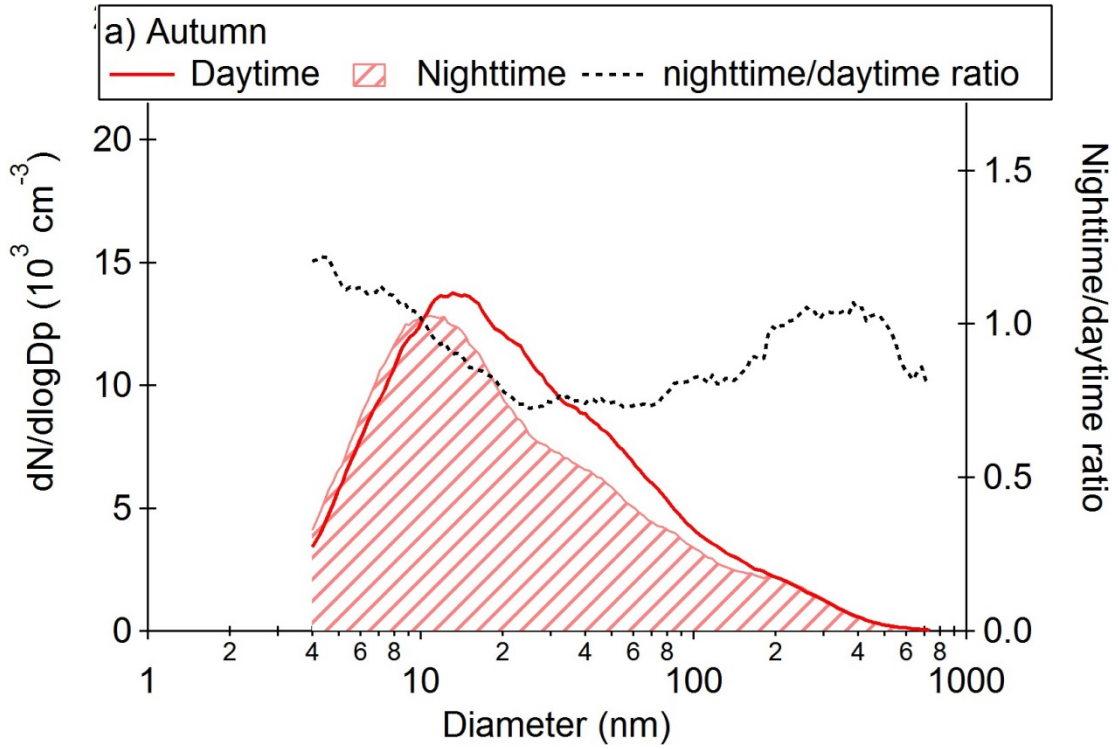
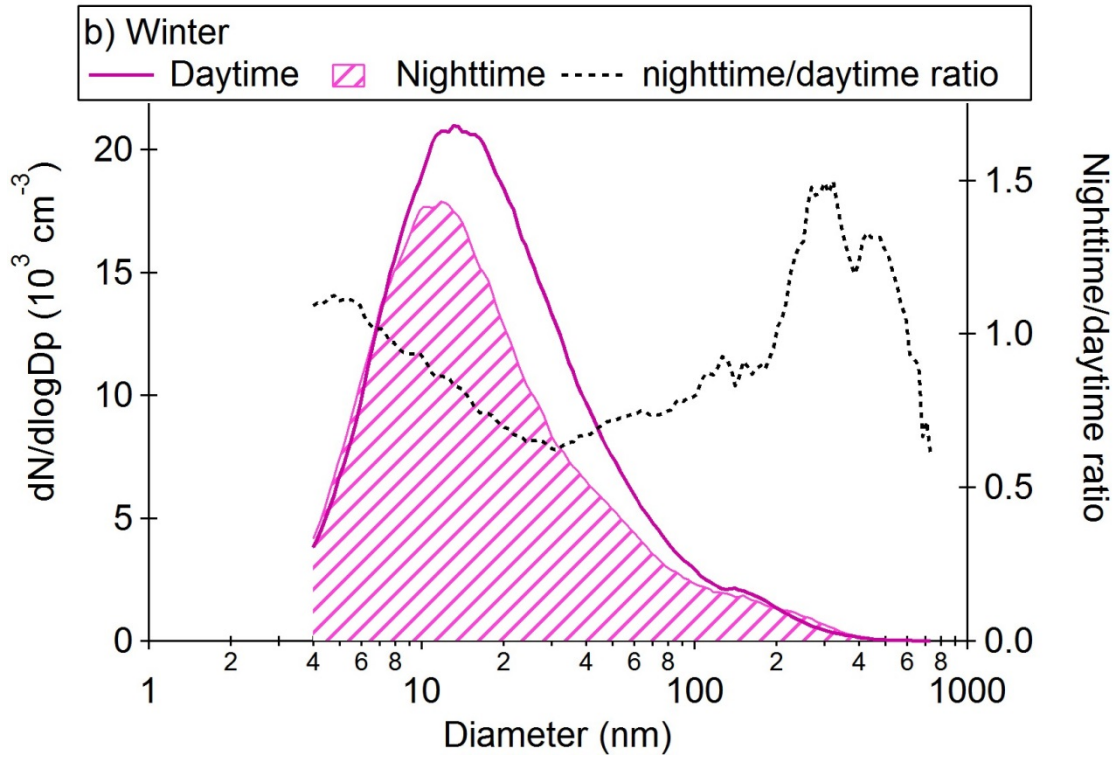


Figure 2. Size distribution of particle number (upper panel), surface and volume (lower panel) concentrations measured in **(a)** autumn, **(b)** winter, **(c)** spring and **(d)** summer (by curve fitting).

Figure 3. Seasonal average concentration and composition of **(a)** ultra-fine (UFPs) and **(b)** sub-micron (PM_1) particles observed at the TARO in Taipei, Taiwan from autumn 2012 to summer 2013.





886
887
888
889
890
891
892
893

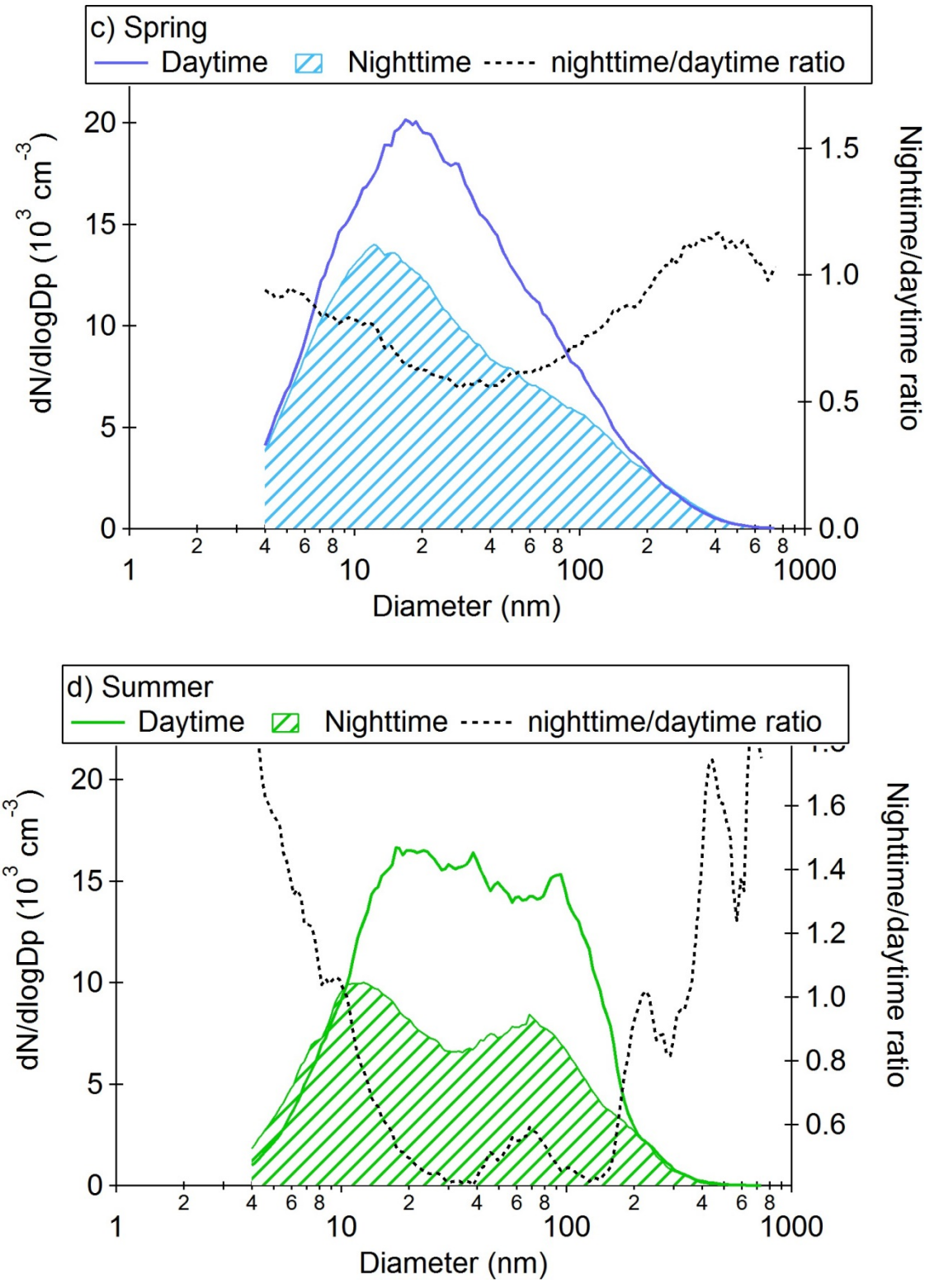


Figure 4. Median PSDs measured during the daytime (07:00 – 17:00 LT) and nighttime (17:00 – 07:00) in **(a)** autumn, **(b)** winter, **(c)** spring and **(d)** summer.

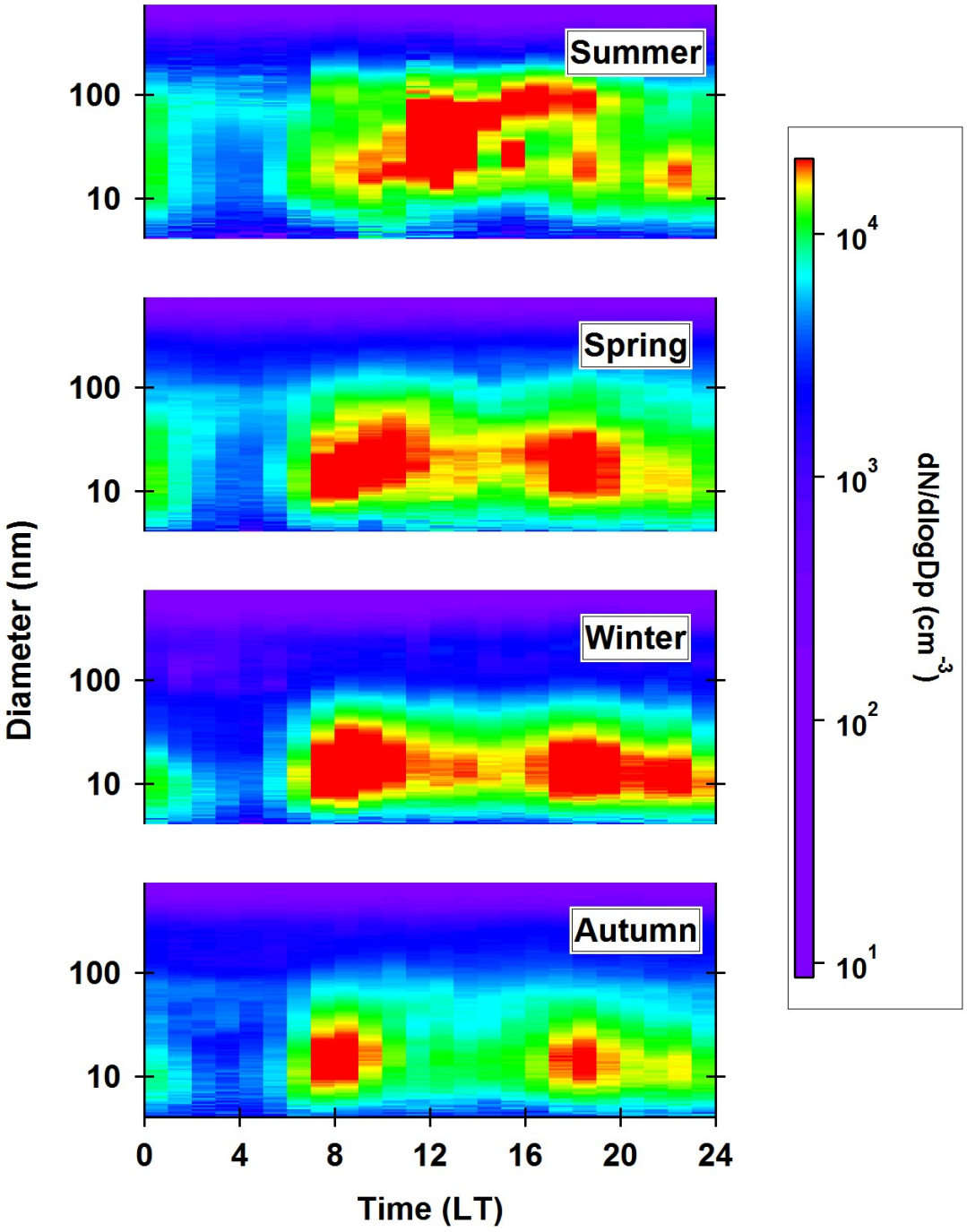
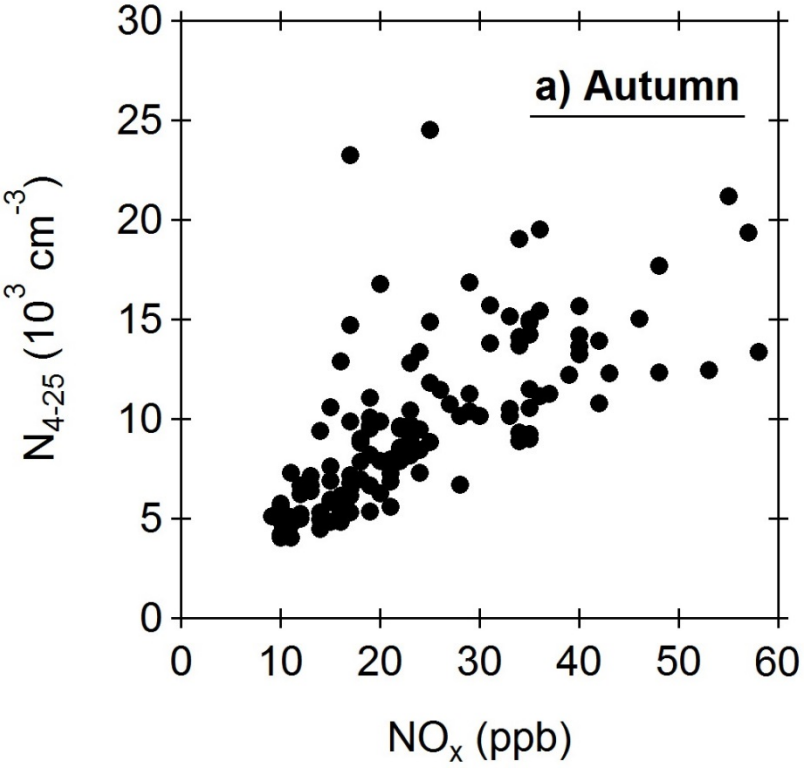
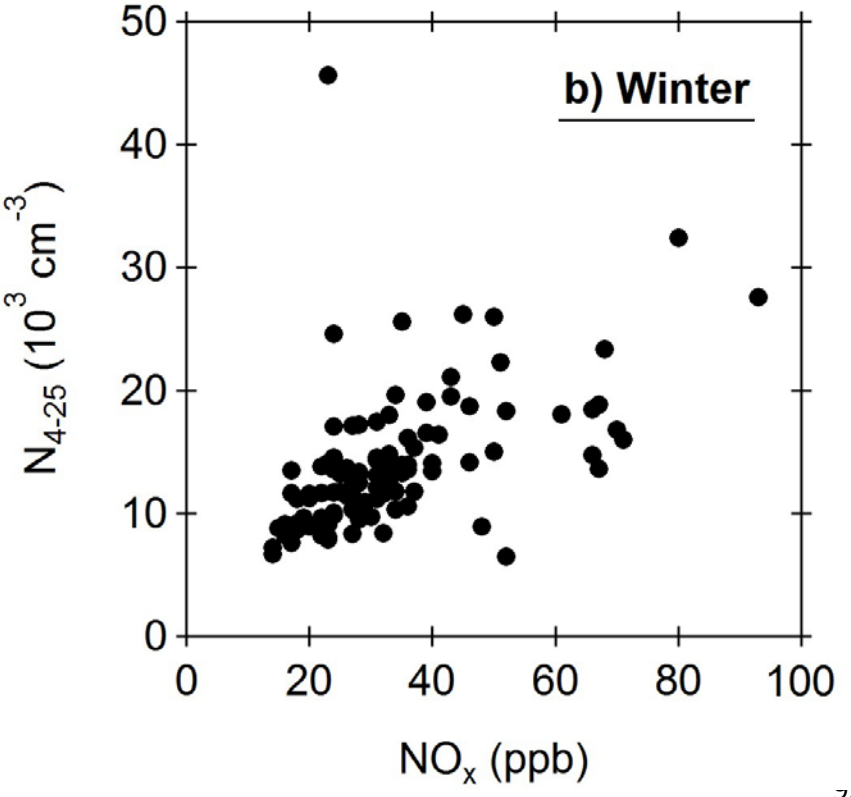


Figure 5. Diurnal variation of particle number size distribution in each season. From lower panel to top panel: autumn, winter, spring and summer.

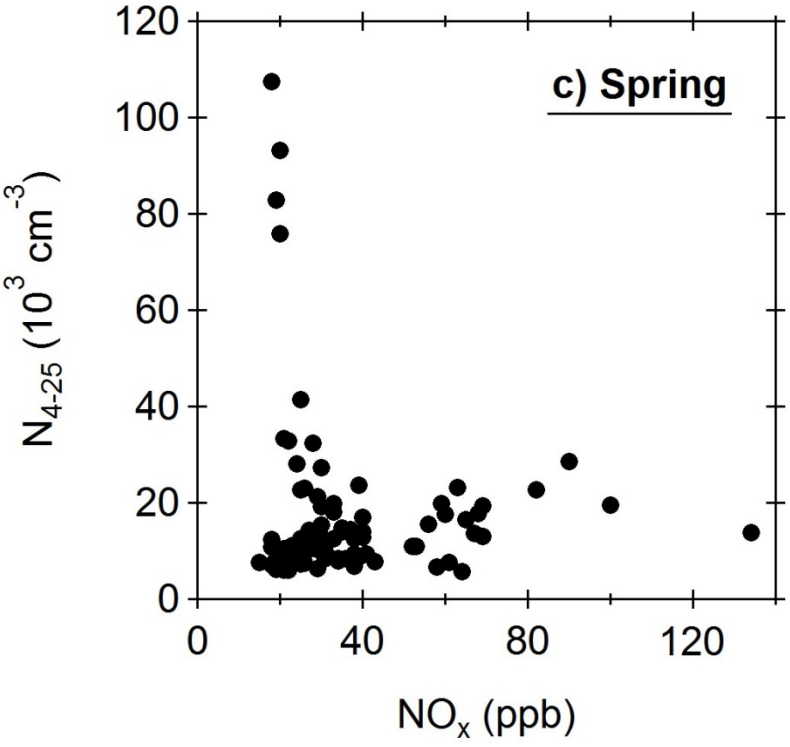


932

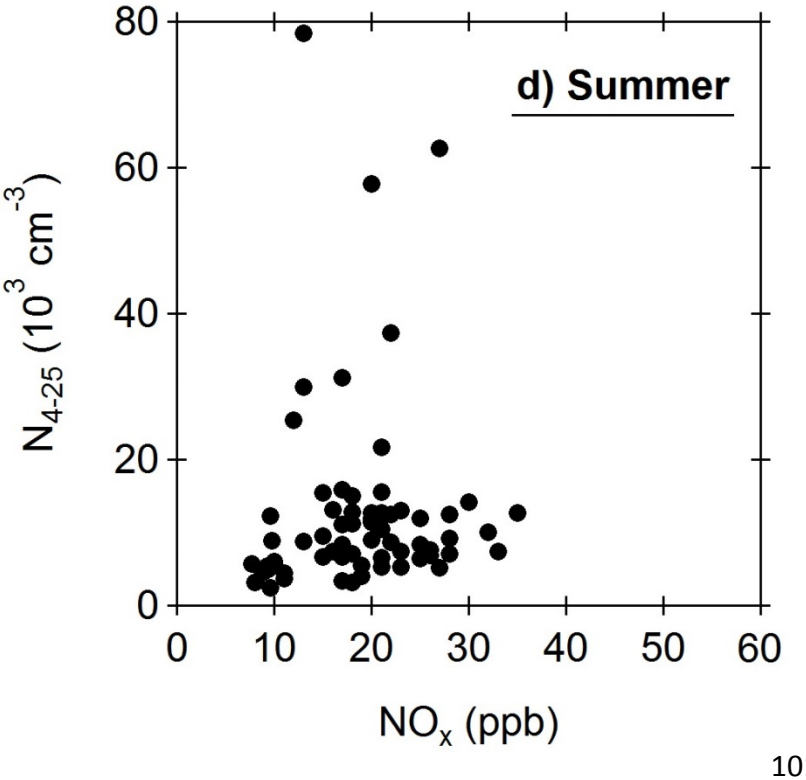


938

959
960
961
962
963



988



1013

1014 **Figure 6**. Scatter plots between hourly $N_{4\text{-}25}$ and NO_x observed in **(a)** Autumn, **(b)** Winter, **(c)**
1015 Spring and **(d)** Summer at TARO site during the period of 07:00 – 17:00 LT.

1016

1017

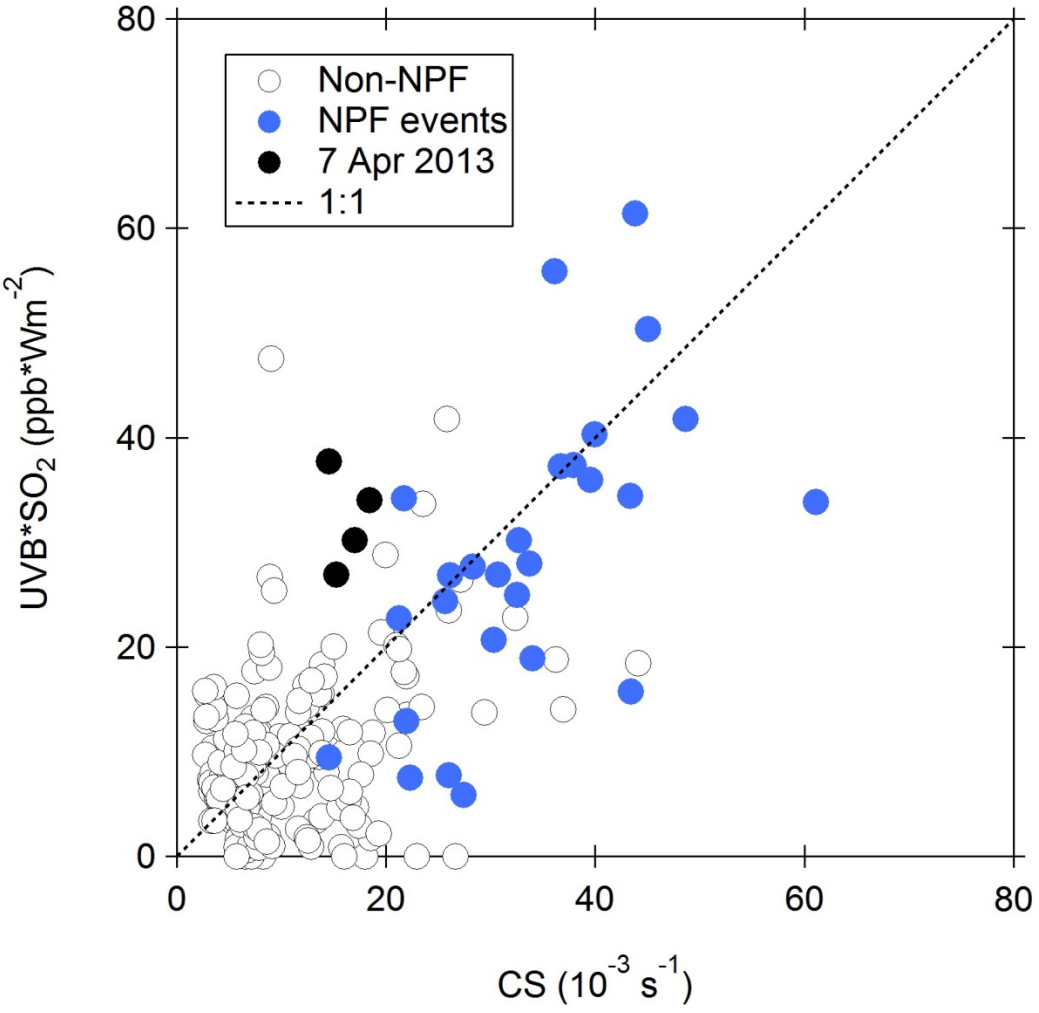


Figure 7. Scatter plot of hourly averaged UVB*SO_2 versus condensation sink at noontime (10:00 – 14:00 LT).

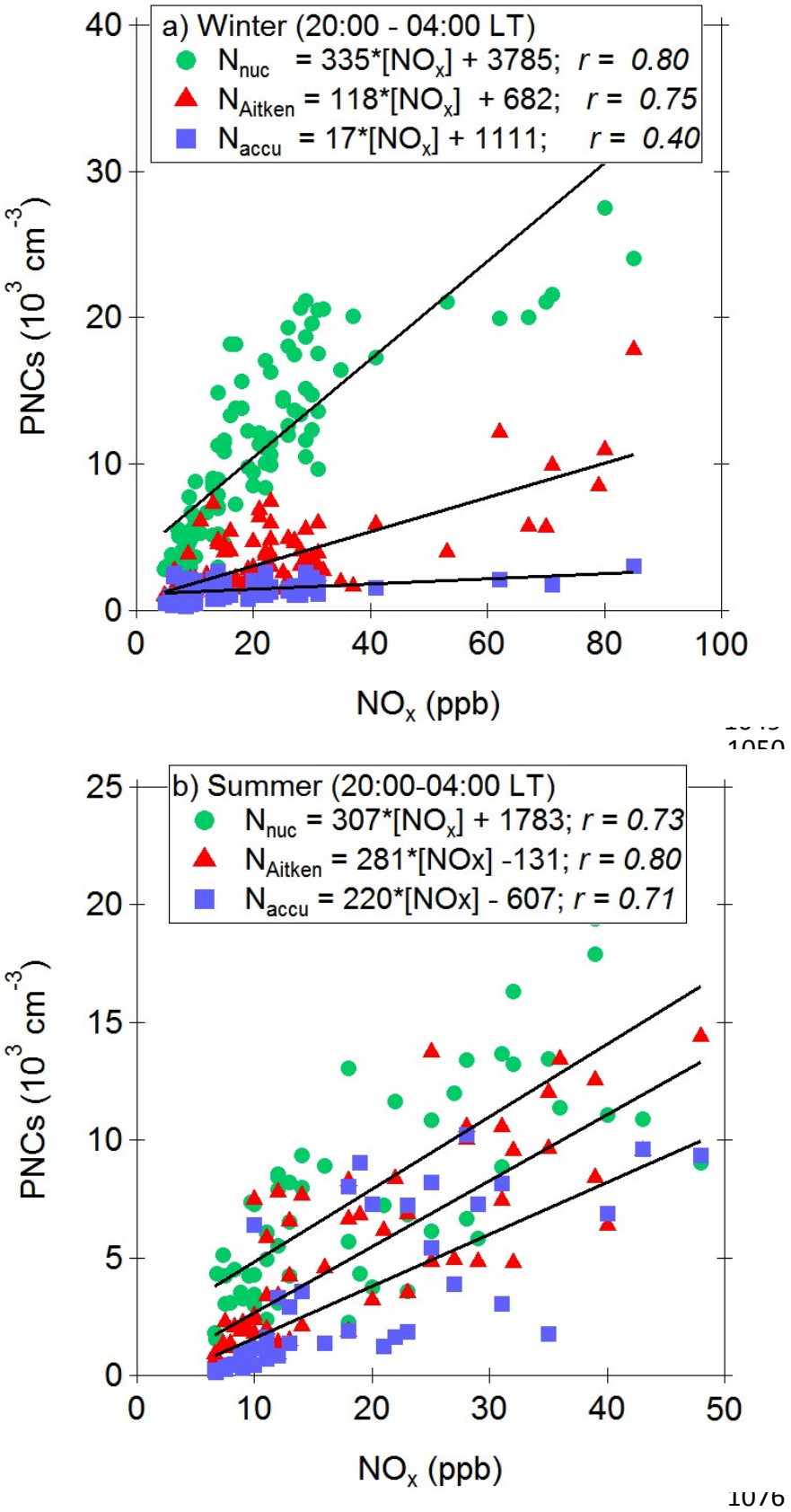


Figure 8. Scatter plots for hourly averaged PNCs vs. NO_x measured during the time period of 20:00 – 04:00 (LT) in **(a)** winter and **(b)** summer, with classification of various particle size ranges.

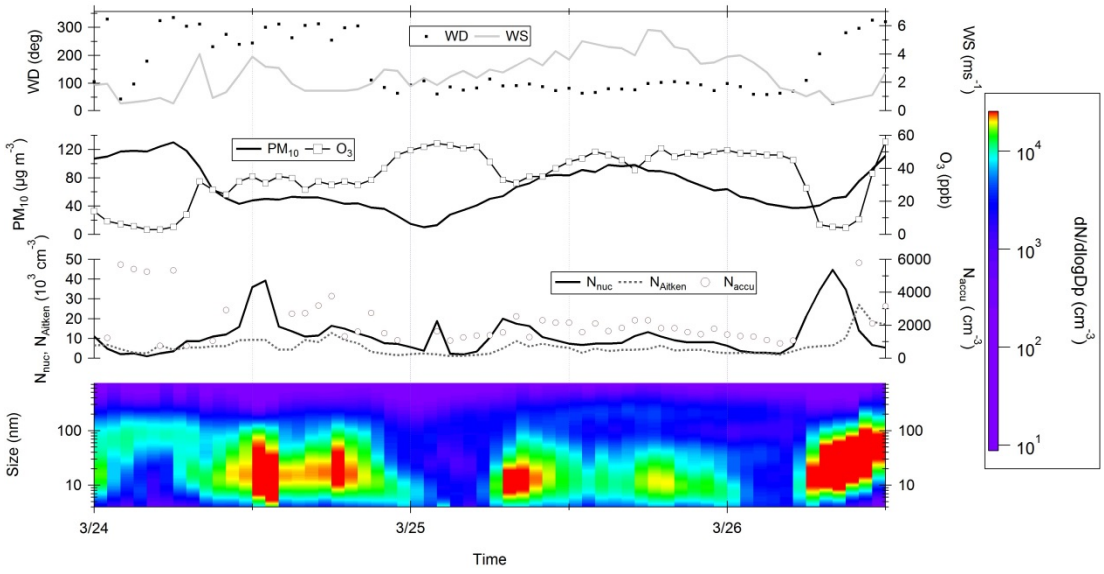


Figure 9. Time series of PSD, the $N_{4\text{-}25}$, $N_{25\text{-}100}$, $N_{100\text{-}736}$, PM_{10}, ozone (O_3) and wind direction/speed measured from 24 - 26 March 2013 (from bottom to top).

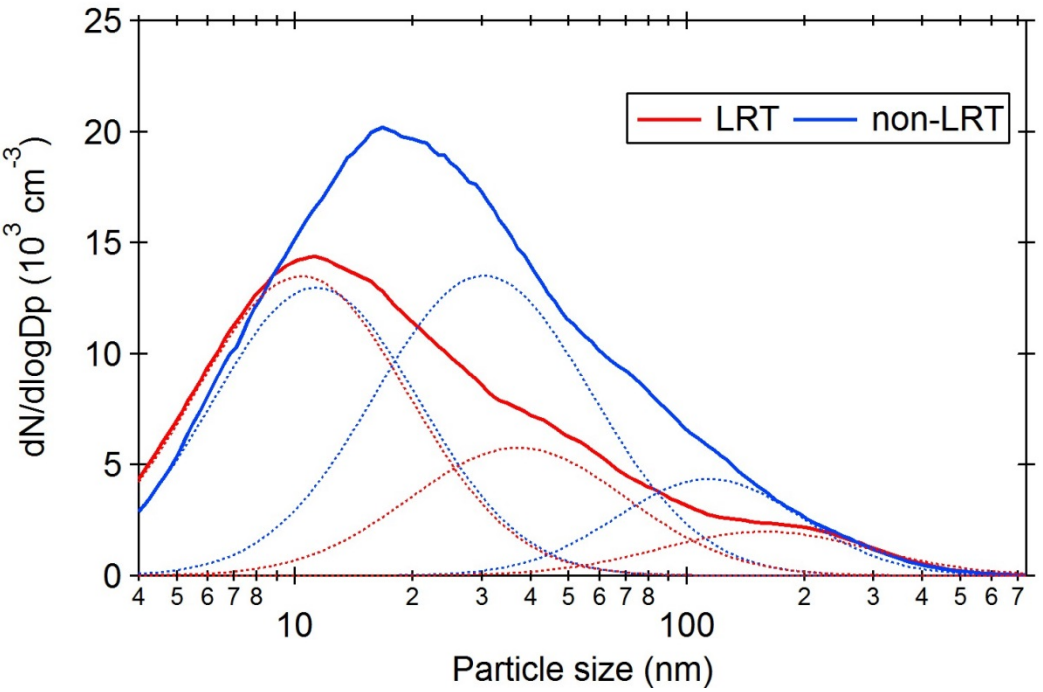


Figure 10. Averaged PSDs for LRT and non-LRT episodes measured during the seasons of winter monsoons. Dashed lines illustrate the PSD of each individual mode.