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Observations of new aerosol particle formation in a tropical urban atmosphere

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HIGHLIGHTS

- New particle formation (NPF) was observed in Singapore in the presence of high SO_2 concentrations.
- NPF events occurred mainly in the afternoon in the presence of high sunlight.
- Meteorological parameters and pre-existing aerosol concentrations had a major influence on the rate of NPF and its intensity.
- Local bush fires were observed to suppress the NPF.

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ABSTRACT

Particle number concentrations (PNC) and particle size distributions (PSD) in the size range of 5.6–560 nm were measured in Singapore during South West (SW) and North East (NE) monsoon periods. The field study was conducted from 27 July 2008 to 15 August 2008 and from 21 January 2009 to 22 February 2009. A distinct peak of PNC in the afternoon was observed in addition to morning and evening rush hour peaks during the SW monsoon period. Concurrent measurements of PSD, SO_2, Black Carbon (BC) and proxy H_2SO_4 concentrations revealed that the afternoon peaks observed during the SW monsoon period were likely due to new particle formation. These nucleation events were frequently observed during the SW monsoon period, but were rarely seen during the NE monsoon period. The effect of meteorological parameters viz. Temperature (T), Relative Humidity (RH), Incoming Solar radiation (SR) on the rate and intensity of nucleation was examined. Strong nucleation events were observed in the presence of high H_2SO_4 concentrations at high T, high SR, and low RH. The newly formed particles did not show any signs of growth during the nucleation events. New particle formation (NPF) events appear to be mainly induced by SO_2 emissions from the local point sources (e.g. petroleum refineries), so when winds blew from that direction nucleation events were prominent. Local bush fires were observed during the course of air sampling due to a prolonged dry spell in the months of January and February 2009. During the occurrence of the local smoke haze induced by bush fires, nucleation events were suppressed.

1. Introduction

Atmospheric aerosols, especially ultrafine particles (UFPs, diameter < 100 nm), play a key role in affecting human health (Nel, 2005; Peters et al., 1997) and can modify climate at local and global scales (IPCC, 2007; James et al., 1997). It was reported that the uncertainties in the Earth's radiation budget caused by anthropogenic changes are mainly due to atmospheric aerosols (IPCC, 2007; Charlson et al., 1992). These aerosols are introduced to the atmosphere by anthropogenic sources such as vehicular traffic and industry and natural sources such as forest fires, dust, sea spray, and volcanoes. Apart from these direct sources, aerosol particles are also formed in the atmosphere through gas-to-particle conversion; this phenomenon is often termed as atmospheric nucleation or new particle formation (NPF). Over the last few decades, atmospheric nucleation has gained increasing attention because of the possible effects of these particles on climate, especially on the number of cloud condensation nuclei (CCN) and the resulting effects on cloud physical and optical properties (Sitaras and Siskos, 2008; Spracklen et al., 2008; Pierce and Adams, 2009). In view of

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these concerns, research is conducted by a number of groups on NPF in various environments ranging from unperturbed rural to heavily polluted urban areas, and from coastal to continental environments (Kulmala et al., 2004; Manninen et al., 2010; Neitola et al., 2011; Shen et al., 2011; Asmi et al., 2011; Boy et al., 2008; Vakkari et al., 2011; Place et al., 2010). A number of studies on atmospheric nucleation have been conducted in coastal environments (O'Dowd and Hoffman, 2005 and references cited there in; Vaattovaara et al., 2006). From these studies, it became clear that the clean air arriving from the open ocean enhances NPF. NPF is regularly observed at continental background environments, including boreal forest (e.g., Mäkelä et al., 1997; Dal Maso et al., 2005), broad-leafed forest (Pryor et al., 2011) and savannah (Vakkari et al., 2011) environments. Vakkari et al. (2011) conducted their study in semi-clean South African savannah environment and observed that 69% of days in an 18 month campaign showed NPF and growth events, while 14% of total sampling days had nucleation bursts of non-growing particles. The large background concentration of aerosols in polluted locations appears to suppress NPF. However, NPF has still been observed in several polluted environments (e.g., Birmili and Wiedensohler, 1998; Birmili et al., 2001; Wehner et al., 2004; Jeong et al., 2004; Yue et al., 2011). Wehner et al. (2004) monitored PNC and PSD in a heavily polluted urban site of Beijing, continuously for 45 days (March 05 to April 18, 2004). They observed NPF events only when particle surface area concentrations were below a critical value (100–2000 $\mu m^2\ cm^{-3}$). Jeong et al. (2004) also observed nucleation events in the urban atmosphere of Rochester, New York. They observed a strong association between the occurrence of nucleation events and elevated SO_2 concentrations. In addition to the usual urban traffic, local emissions from a thermal power plant were also observed to influence the atmospheric concentration of SO_2 at the sampling area.

The mechanisms involved in NPF are still incompletely understood. Several mechanisms have been proposed such as binary nucleation of water-sulfuric acid (H_2SO_4) (Kulmala and Laaksonen, 1990), ternary nucleation of H_2O–H_2SO_4–ammonia (NH_3) (Merikanto et al., 2007; Nepari et al., 2002) and ion-mediated nucleation (Yu, 2006). Efforts are also underway to provide insights into nucleation mechanisms by analyzing observed nucleation events from diverse environments with respect to the concentrations of potential precursors. Such studies have related the nucleation rate to the concentration of H_2SO_4 (Weber et al., 1997; Sihto et al., 2006; Riipinen et al., 2007; Kuang et al., 2008). Laboratory experiments appear to confirm that the presence of H_2SO_4 is critical for nucleation, and have also elucidated the roles of ammonia (Kirkby et al., 2011) and organics (Metzger et al., 2010; Kiendlar-Scharr et al., 2012 Pierce et al., 2012) in NPF. Moreover, biogenic emissions of iodine vapors from sea could also play a dominant role in nucleation events that are observed in coastal environments (O'Dowd and Hoffman, 2005). Most of the studies reported in the literature were conducted in the temperate zones where distinct and definitive seasonal variations in atmospheric conditions can be observed (Kulmala et al., 2004). However, there is a lack of field-based investigations in tropical regions close to the equator where the climatic conditions show little variations throughout the year, and the prevalent sunny and clean atmosphere may be favorable for nucleation events.

In order to expand the existing knowledge-base on atmospheric nucleation, we measured the number concentrations and size distributions of UFPs in Singapore during two climatologically different periods, SW and NE monsoons. BC and SO_2 concentrations along with key meteorological parameters were also measured. The study was conducted with the objective to investigate atmospheric nucleation events within a tropical urban environment and their possible dependence on prevailing meteorological conditions. During the course of this study, local bush fires took place and the effect of these fires on the PNC and PSD was also studied. This work represents the study of its kind in investigating NPF events in the tropical region of South East Asia where biomass burning episodes frequently take place.

2. Experimental methods

2.1. Sampling site

Measurements of the number, mass concentrations and size distribution of airborne particles were conducted from 27 July 2008 through 15 August 2008 and from 21 January 2009 to 22 February 2009 at the Atmospheric research station located in the National University of Singapore campus (latitude: 1° 18′ N; longitude: 103° 66′ E). The two sampling periods represented South-West (SW) and North-East (NE) monsoons, respectively; these are the two prevalent monsoons in the tropical South-East Asia. Air sampling was conducted on the roof top of one of the tallest buildings at the Faculty of Engineering at an altitude of 67.0 m above sea level with no visible obstructions. The sampling site (Fig. 1.) is considered to be an urban location influenced by vehicular traffic; a busy expressway connecting to the Central Business District runs toward South East approximately at a distance of 200 m north from the sampling location and also another road runs southward at a distance of 50 m west from the sampling site. Petroleum, petrochemical and specialty chemical industries are located at a distance of 5–10 km to the South West. Apart from the vehicular and industrial emissions, the site is also influenced by sea spray and emissions from ships as the sampling site is approximately 1 km from a busy coastal port.

2.2. Measurements and methods

Meteorological parameters (temperature (T), pressure (P), relative humidity (RH), wind speed (WS), direction (WD), solar radiation (SR), and precipitation) were acquired every 5 min by an automated meteorological station located at the sampling site. P, T, RH were measured using suitable sensors (Vaisala, Model CS 500), WS & WD using anemometer and wind vane (RM Young, Model 03001). SR was measured using Pyranometer (LI-COR, Model LI 200X). The entire system was connected to a Console/Receiver (Part Number 6310) and to a personal computer via Weather Link for Vantage Pro Data Logger (CSI, Model CR10X).

Fast Mobility Particle Sizer (FMPS, Model 3091d, TSI Inc.) was employed at the same location to measure the PNC and PSD concurrently over a wide size range. Mobility diameter in the range of 5.6–560 nm was scanned by the FMPS every second continuously throughout the sampling period and was recorded using a personal computer connected to it at the sampling location. At the beginning of the field study, frequent instrumental failures were encountered due to incompatibility between the personal computer used for data acquisition and the software needed for recording the samples and the data gathered during that period were discarded.

BC concentrations were measured at five minute intervals from 6 August 2008 to 15 August 2008 and 2 February 2009 to 25 February 2009 using an Aethalometer (Magee Scientific Inc.). Sulfur dioxide (SO_2) concentrations were also measured during the same time period using pulsed fluorescence SO_2 analyzer (Model 45C, Thermo Scientific) at an interval of 1 h.

Sulfuric acid concentration was estimated with a proxy using the following equation described by Petaja et al. (2009)

$$H_2SO_4\ \text{proxy} = k_3 \frac{[SO_2] \times SR}{CS} \quad (1)$$

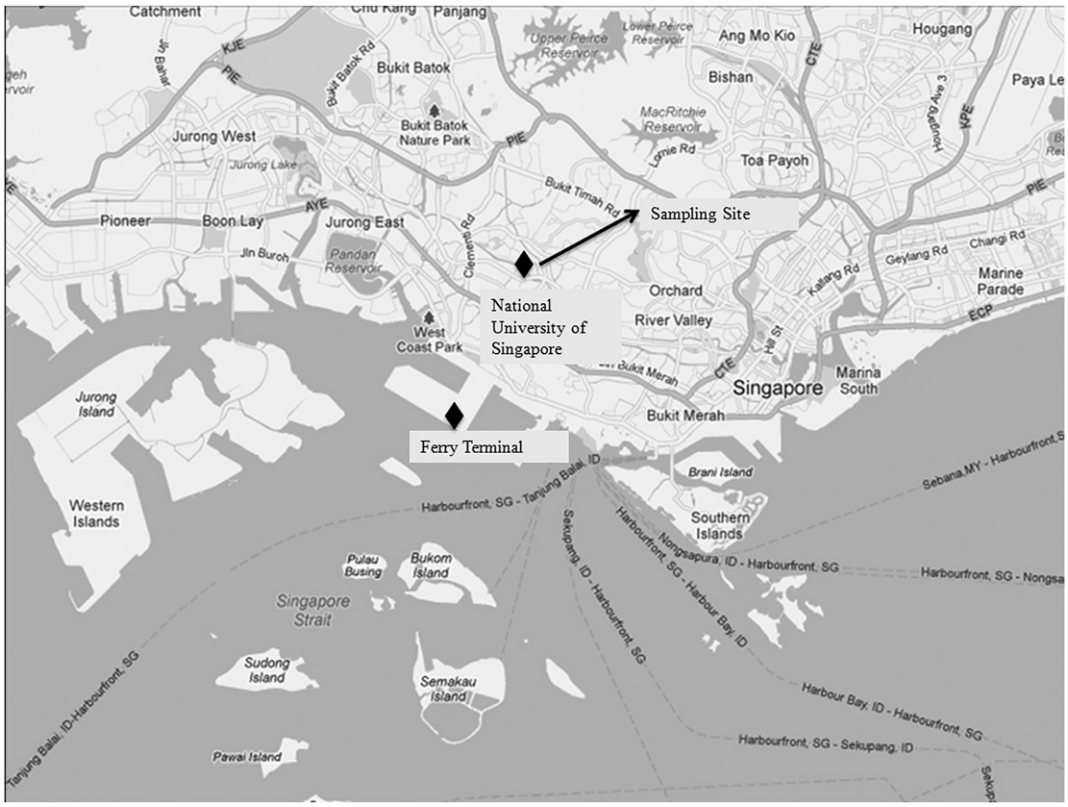

Fig. 1. Sampling location in Singapore.

where SO_2 is the measured sulfur dioxide concentration in molecules cm^{-3}, SR is solar radiation in W m^{-2}, CS is the condensation sink in s^{-1}, calculated from the aerosol particle size distribution measured using FMPS, and the empirical scaling factor, k_3, as given by Equation (2) (Petaja et al., 2009)

$$k_3 = 1.4 \times 10^{-7} \times SR^{-0.7} \quad (2)$$

H_2SO_4 proxy estimation was developed based on the measurements in boreal forests and therefore the absolute values may not be applicable to the Singapore tropical conditions. Despite this drawback, these estimations still provide valuable insights into the H_2SO_4 influence on new particle formation.

3. Results and discussion

3.1. Meteorology

Statistical characteristics of meteorological parameters during the two periods are shown in Table 1. It was observed that there is no significant difference (Student's *t*-test, $p > 0.05$) in *P*, *T*, RH and SR during both SW and NE monsoon periods. This is typical in tropical countries where the climate conditions show only little variations throughout the year. The only significant difference noted between the two periods was in the wind speed and direction (Fig. 2). During the NE monsoon, the mean wind speed was higher ($p < 0.05$) than that of the SW monsoon period. The winds were mostly from NE during the entire sampling period in the NE monsoon. However, during the SW monsoon the winds blew from different directions ranging from South East to South West and also the percentage of low (<0.5 m s^{-1}) wind speeds (20.7%) was relatively higher during the SW monsoon than that during the NE monsoon period (12.5%). In addition, the collision of strong see breezes from both the East and West directions of Singapore leads to strong convective winds, especially during the afternoon hours (Joseph et al., 2008).

3.2. Particle number concentration

Statistical parameters of PNC measured in the three size ranges (5.6–50 nm; 50–100 nm; 100–560 nm) during the two different sampling periods (SW and NE monsoon) are shown in Table 2. Significantly higher ($p < 0.05$) total PNC were observed during the SW monsoon compared to the NE monsoon period. Among the three size ranges, airborne particles in the size range 5.6–50 nm did not show any significant difference ($p > 0.05$) between the two periods. However, particles with diameter in the range of 50–

Table 1
Statistical summary of meteorological parameters measured during SW and NE monsoon periods.

	Pressure (hPa)	Air temp (°C)	RH (%)	Wind speed (m s^{-1})	Wind direction (°)	Solar radiation (W m^{-2})
SW monsoon						
Mean	1001	27.6	72.8	1.7	144.8	185.9
Median	1001	27.8	73.0	1.7	142.8	202.1
SD	0.6	0.5	2.2	0.5	15.5	42.0
Minimum	999	26.2	68.3	0.7	117.4	89.4
Maximum	1002	28.0	77.1	2.6	184.6	223.9
NE monsoon						
Mean	1002	26.3	72.6	2.8	74.0	162.2
Median	1002	26.1	71.6	2.9	61.8	158.6
SD	1	0.7	3.8	1.0	35.3	35.2
Minimum	1000	25.4	65.1	1.0	42.0	101.1
Maximum	1003	28.4	80.0	4.4	183.0	229.9

SD: Standard deviation.

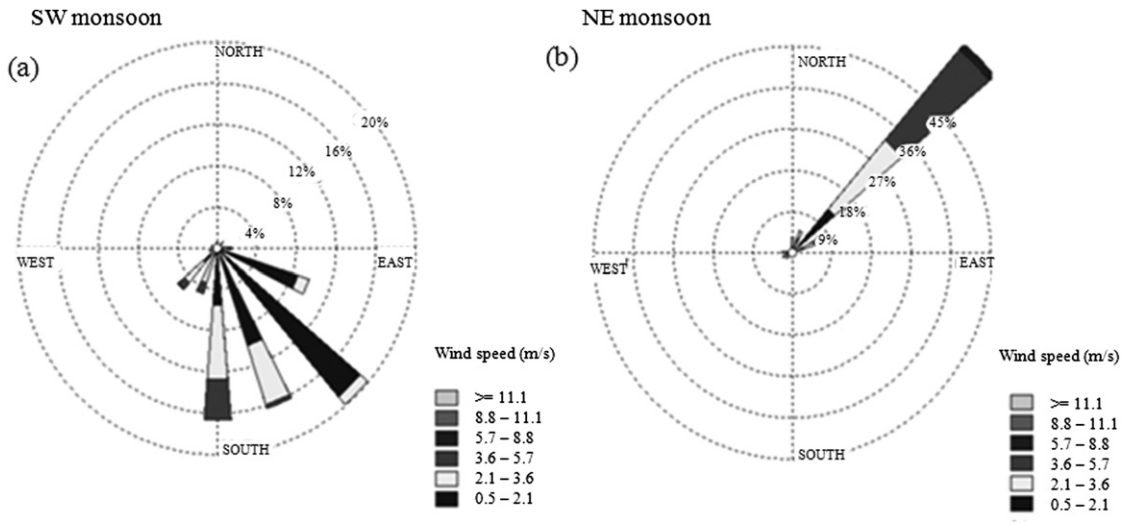

Fig. 2. Wind rose diagrams during the two sampling periods (a) NE monsoons (b) SW monsoons.

100 nm and 100–560 nm showed a significantly higher concentration ($p < 0.05$) during the SW monsoon period compared to the NE monsoon period. The variation (indicated by standard deviation (SD)) of PNC for all size fractions measured was higher during the NE monsoon period than that in the SW monsoon period. The SW monsoon was dominated by mild (0.5–2.1 m s^{-1}) and very low winds (<0.5 m s^{-1}) whereas during the NE monsoon weather was more turbulent with the wind speeds reaching about 8 m s^{-1} (see Fig. 2). Due to the turbulent nature of the atmosphere, the dispersion and dilution of the atmospheric gases and particulates was generally greater during the NE monsoon, likely contributing to lower PNC and higher variability in their concentrations. During the SW monsoon period, winds blew over petrochemical industries, open sea and the seaport that are located to the South and South West of the sampling site. Emissions from these sources along with emissions from traffic sources contribute to the high PNC that is observed during this period. During the NE monsoon period, winds from the NE meant that the site was not influenced by petrochemical or ship emissions, but by the local vehicular traffic.

3.3. Diurnal variations

The diurnal variations of PNC, BC, SO_2, and proxy H_2SO_4 measured during the two sampling periods are illustrated in Fig. 3. During the SW monsoon period (Fig. 3a), three distinguishable peaks were observed in the diurnal variation of PNCs, one in the morning, one in afternoon and the other in the evening. The morning PNC peak usually occurred between 08:00 and 09:00 with a value of 17,523 ± 1337 cm^{-3} (mean ± SD). The afternoon peak usually occurred at 14:00 with a value of 22,633 ± 1993 cm^{-3} while the evening peak occurred between 17:00 and 19:00 though sometimes as late as 20:00 with PNC ranging between 19,892 ± 2337 cm^{-3} at 17:00 h and 19,423 ± 1341 cm^{-3} at 19:00. Morning and evening PNC peaks occurred at the same time as rush hour traffic and are most likely associated with traffic emissions (see analysis of BC concentrations below). However, the daily maximum PNC was observed during the afternoon despite the fact that traffic was sparse at that time, suggesting that the afternoon peak may not be associated with traffic emissions. Previous studies in the urban atmosphere of Rochester (Jeong et al., 2004), and Helsinki (Laakso et al., 2003) showed similar trends in the diurnal variation of number concentrations. In contrast, during the NE monsoon period, there were no afternoon peaks of PNC (Fig. 3b); only morning and evening peaks were observed with PNC of 15,556 ± 1265 cm^{-3} and 19,424 ± 3624 cm^{-3}, respectively.

Table 2
Statistical summary of hourly averaged particle number concentration (# cm^{-3}) during the two sampling periods.

Diameter (nm)	5.6–50	50–100	100–560	5.6–560
SW monsoon				
Mean	9.84×10^3	5.41×10^3	1.78×10^3	1.70×10^4
Median	8.69×10^3	4.60×10^3	1.65×10^3	1.56×10^4
SD	5.29×10^3	2.67×10^3	6.14×10^2	7.16×10^3
Min	1.36×10^3	1.34×10^3	3.78×10^2	3.33×10^4
Max	4.94×10^4	1.56×10^4	4.33×10^3	5.48×10^4
NE monsoon				
Mean	1.01×10^4	3.09×10^3	1.56×10^3	1.47×10^4
Median	8.05×10^3	1.53×10^3	1.21×10^3	1.12×10^4
SD	7.21×10^3	3.99×10^3	9.88×10^2	1.02×10^4
Min	1.71×10^3	9.16×10^1	3.85×10^2	3.18×10^3
Max	7.00×10^4	2.45×10^4	6.55×10^3	7.72×104

SD: Standard deviation, Min: Minimum, Max: maximum.

Analysis of concentrations of BC and SO_2 can lead to additional insights into the likely sources of particles. Vehicular traffic was likely to be an important source of airborne particles measured at the study site as revealed by high concentrations of BC in the urban atmosphere (Fig. 3). BC concentrations peaked in the morning (09:00) and evening (18:00) corresponding to traffic rush hours and were very low in the afternoon (14:00–16:00) during which the local traffic was sparse regardless of the monsoon periods. However, diurnal variations of SO_2 were similar to those of PNC during both the measurement periods. SO_2 concentrations during the SW monsoon period were relatively higher (4.5 μg m^{-3}; 0.2–4.6 ppb) compared to the NE monsoon period (0.6 μg m^{-3}; 0.2–0.4 ppb) and peaked in the afternoon (14:00–16:00) along with the morning and evening rush hour peaks (Fig. 3). The main sources of SO_2 are likely to be the petrochemical industry located in the SW direction of the sampling site in addition to emissions from shipping traffic (about 3300 ships transecting Singapore per day). When the wind direction shifted to the NE sector, SO_2 concentrations decreased at the PSD measurement site due to the lack of large SO_2 sources in this direction. The occurrence of the afternoon PNC peak during the SW monsoon appears to be related to the observed increase in the SO_2 concentration during the same time. While the BC concentration was low in the afternoon (14:00–16:00) with a mean value of 854 ng m^{-3}, the PNC and SO_2 concentrations peaked

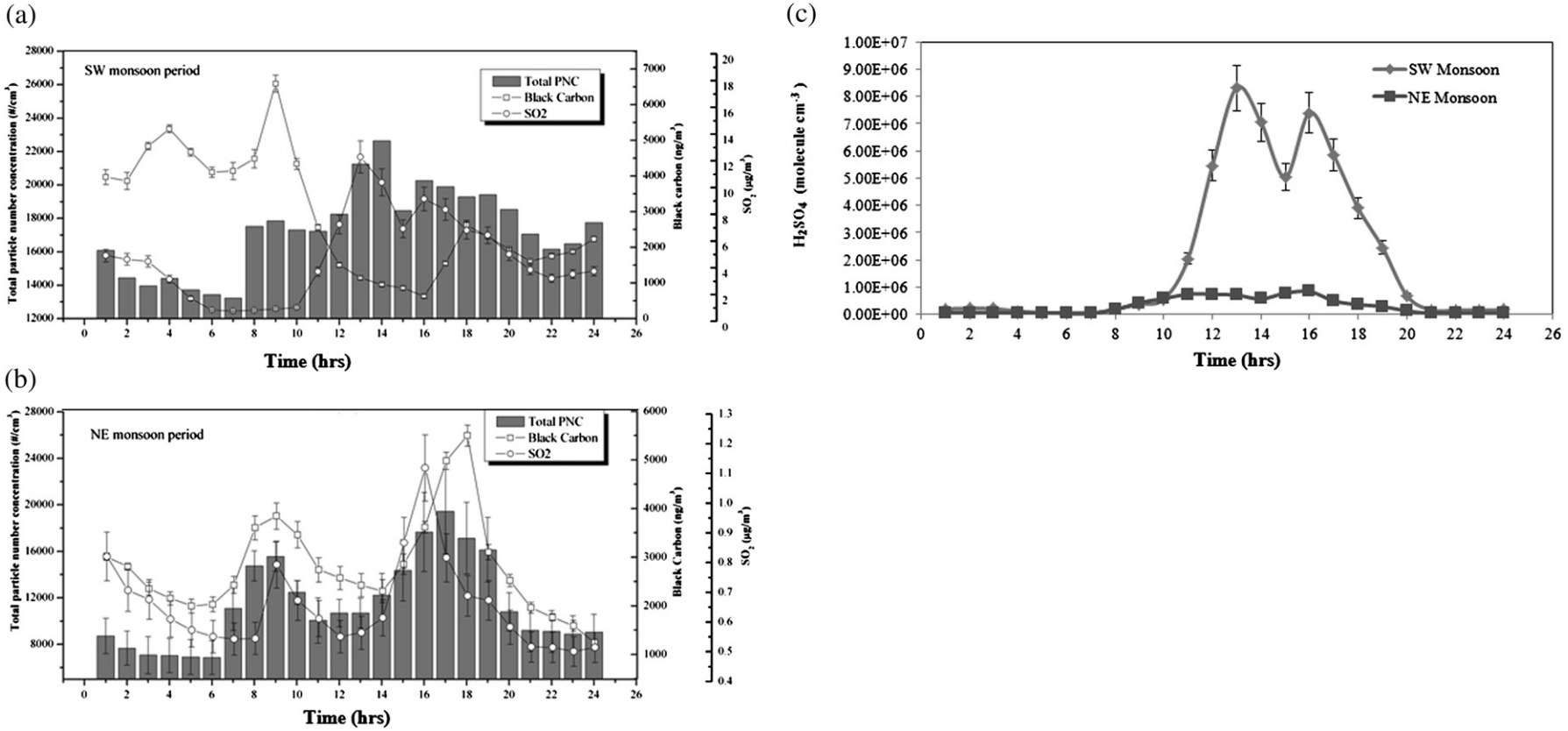

Fig. 3. Diurnal variations of hourly average particle number concentrations, black carbon, SO_2, proxy H_2SO_4 during SW and NE monsoon periods in Singapore.

(22,633 ± 1993 cm^{-3} and 12 ± 1 μg m^{-3}) suggesting that the afternoon peak was not related to BC, but due to new particle formation (NPF) linked to SO_2. It has been widely reported that gaseous SO_2 is closely associated with the occurrence of NPF (e.g. Menon et al., 2002; Kulmala et al., 2004; Kuang et al., 2008). The concentrations of H_2SO_4 proxy (Fig. 3c) further confirm the formation of new particles in the afternoon hours during the SW monsoon period. The H_2SO_4 vapor-phase concentrations were high during the afternoon period consistent with the observations of NPF events that occurred during the afternoon time. Also as shown in the figure, the concentration of H_2SO_4 during the NE monsoon periods was relatively lower compared to that during the SW monsoon.

3.4. Particle size distribution

3.4.1. SW monsoon

The particle size distributions (PSD) observed for morning (08:00–09:00), afternoon (14:00–15:00) and evening (17:00–19:00) peaks in the total PNC during the SW monsoon period are shown in Fig. 4a. The PSD peaked with a diameter of 45.3 nm and a peak concentration of 1760 cm^{-3} and 1486 cm^{-3} in the morning and evening, respectively whereas in the afternoon the PSD peaked at a diameter of 10.8 nm with a peak concentration of 3690 cm^{-3} which was 2–2.5 times higher than that observed in the morning and evening peaks. As discussed earlier, the low BC and high SO_2 and proxy H_2SO_4 concentrations observed during the afternoon periods indicated that the observed particles were unlikely to be directly emitted from combustion sources such as vehicular traffic, but were rather formed in the atmosphere by gas-to-particle (GTP) conversion. The shift in PSD supports the hypothesis that the sudden increase in PNC in the afternoon can be attributed to freshly formed particles through GTP.

Small peaks of nuclei mode particles were also observed in the morning. These particles are likely due to vehicular emissions, as the hot exhaust from combustion engines containing volatile precursors when diluted and cooled tends to form nuclei mode particles (Kittelson, 1998). Jeong et al. (2004) reported similar findings. They found that the nuclei mode particles observed in the morning coincided with the traffic rush hours and the dominant particle size range was between 20 and 100 nm. They attributed the afternoon peaks with a dominating particle size in the range of 11–30 nm to NPF. PSD during the SW monsoon exhibited bi-modal distributions. Log normal fittings were done for both the modes and key parameters such as mean (μ), geometric standard deviation (σ) and total number concentration for the particular mode (N_i) were calculated and are shown in Table 3a.

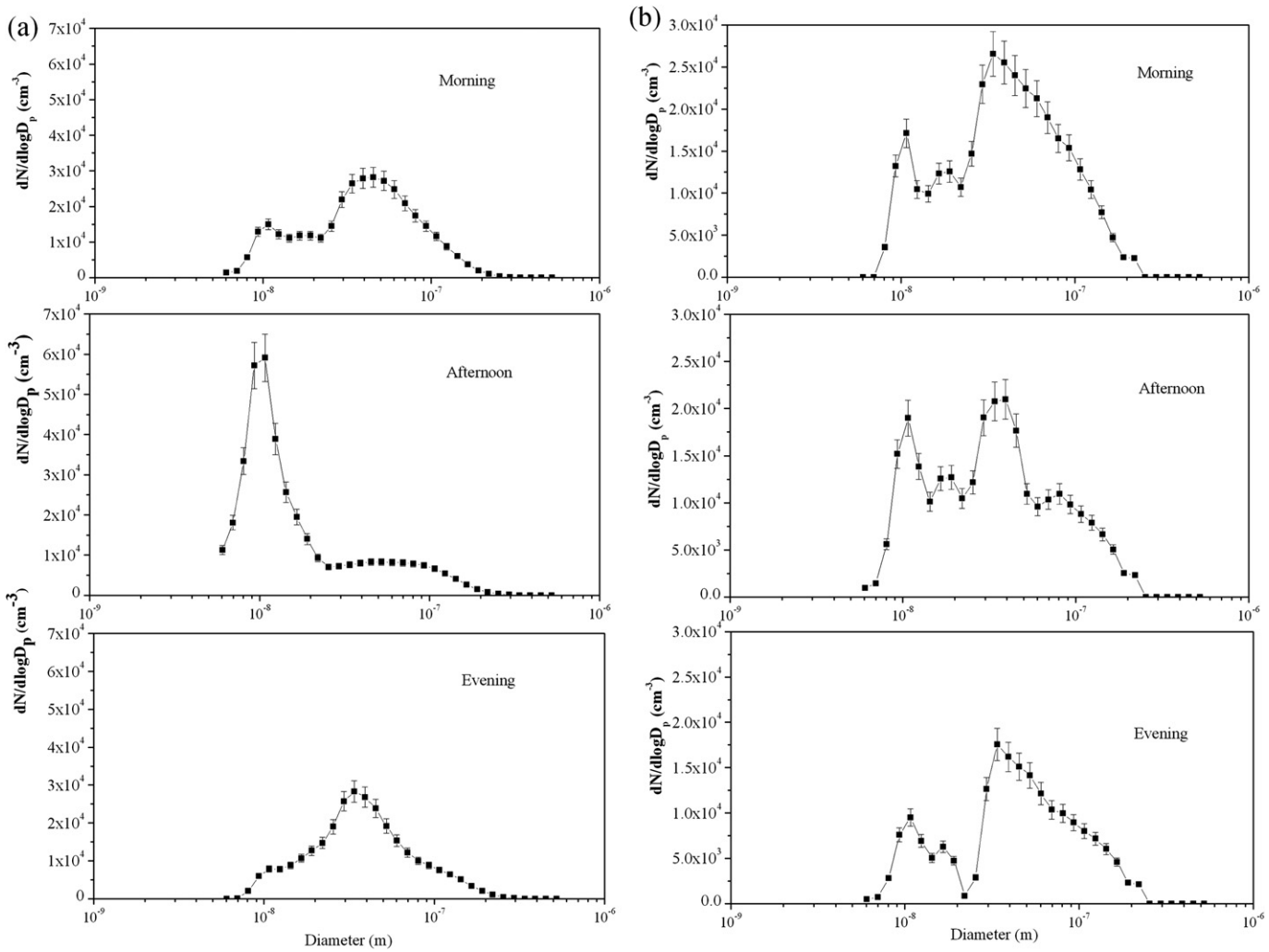

Fig. 4. Particle number size distributions during morning, afternoon, and evening peaks observed in a typical day (a) SW monsoon period (b) NE monsoon period.

Table 3a
Log-normal distribution parameters of particle size distribution during SW monsoon.

SW monsoons	Mode-1			Mode-2		
	N (# cm^{-3})	μ (m)	σ	N (# cm^{-3})	μ (m)	σ
Morning	5933	1.24×10^{-8}	1.4	16,090	5×10^{-8}	1.7
Afternoon	17,906	1.05×10^{-8}	1.4	6226	5.6×10^{-8}	1.8
Evening	4410	1.4×10^{-8}	1.3	13,450	5.4×10^{-8}	1.9

Mode-1 (D_p: 5.6–25 $\times$ 10^{-9} m); Mode-2 (D_p: 25–560 $\times$ 10^{-9} m).

3.4.2. NE monsoons

The PSD observed during the morning, afternoon and evening peaks in the NE monsoon periods was slightly different from the PSD observed during the SW monsoon. As shown in Fig. 4b, morning, afternoon and evening size distributions were bi-modal with peak number concentrations of 861 and 1468 cm^{-3} in the morning, 1108 and 1021 cm^{-3} in the afternoon and 750 and 1092 cm^{-3} in the evening for diameters of 10.8 nm and 34 nm, respectively. Since winds mainly blew from the NE direction over a busy expressway, the particles observed at the monitoring site were likely of vehicular origin and therefore the PSD observed showed little variations throughout the day. The PSD during the NE monsoon exhibited tri-modal distributions. There are three distinct peaks observed in the size distribution and log-normal distributions for all three modes were fitted. The key parameters pertaining to PSD (μ, σ and N_i) for the three modes were calculated and are shown in Table 3b.

3.5. Biomass burning

Events of local bush fires within Singapore occurred during the NE monsoon period. The bush fires took place in a vegetated area along Bukit Batok that lies to the North West of the sampling site (Fig. 1). The outburst of bush fires was due to prolonged hot and dry conditions prevalent in Singapore during the NE monsoon period, February was the driest month of 2009 in Singapore. The prevailing winds transported the emissions to the sample site, increasing the observed particulate loading. Fig. 5a shows the hourly average number concentrations during a normal day and during the event of bush fires. As shown in the figure, the PNC peaked at an hourly average value of 37,800 cm^{-3} in the evening hours, which was almost twice the number concentrations observed in the morning. This evening peak coincided with the timing of bush fires which usually started in the late afternoon driven by very hot and dry conditions. During hazy days, the observed particle size distribution (shown in Fig. 5b) was slightly different from the general trend discussed earlier in the absence of hazy conditions. It was found that the particles peaked at a diameter of 52.3 nm with a peak concentration of 6145 cm^{-3} and the total PNC reaching very high values of 45,000 cm^{-3}. It was also found that the BC concentration also increased to abnormally high values (e.g. 8273 ng m^{-3}), and the BC concentrations remained high for a long period of time.

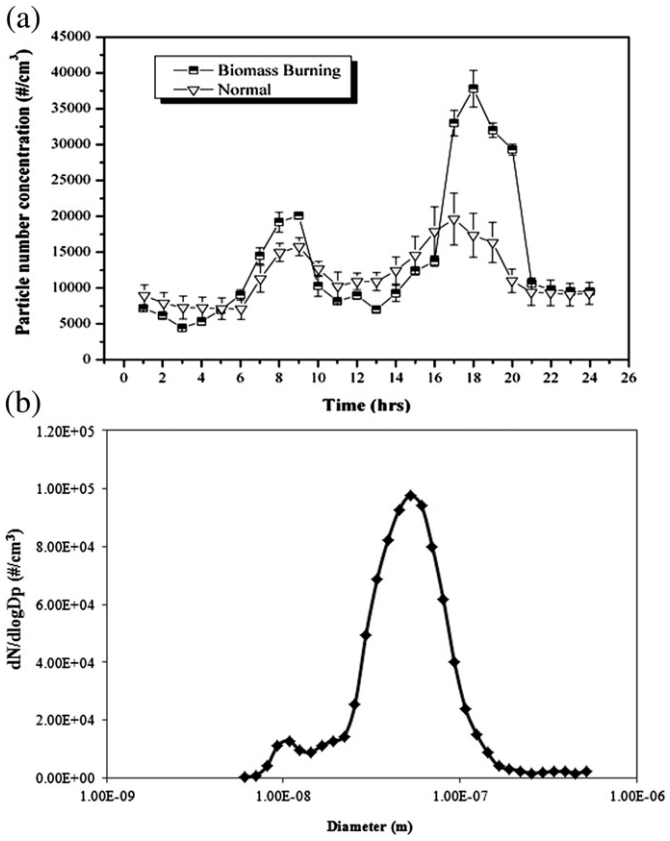

Fig. 5. (a): Hourly average number concentrations of particles observed during the event of bush fires and during a normal day without bush fires in NE monsoon period; (b) Particle size distribution during the event of bush fires.

3.6. Nucleation events

A nucleation event is identified when there is a sudden increase in the number concentrations of airborne particles in the nucleation mode, with sizes less than 25 nm in diameter. The criterion is consistent with the one suggested by Dal Maso et al. (2005). According to Dal Maso et al. (2005), apart from distinctly new mode of particles appearing in nucleation mode size range, the particle formation event should last over a time span of several hours, accompanied by the growth of particles with an increase in particle diameters. However, Dal Maso et al. (2005) described that the latter criteria are applicable only when the new particle formation occurs uniformly over a wide geographical area such as dense boreal forest regions. Fig. 6 shows the representative NPF events observed in this study and the condensation sink (CS) on the respective days; CS was estimated as per the procedure reported by Dal Maso et al. (2005). We observed that on an average, the CS in Singapore is relatively higher (see Tables S1 and S2 in the supplementary section) when compared to most of the studies reported from other parts of the world (Kulmala et al., 2001; Dal Maso et al., 2005; Boy

Table 3b
Log-normal distribution parameters of particle size distribution during NE monsoon.

NE monsoons	Mode-1			Mode-2			Mode-3		
	N (# cm^{-3})	μ (m)	σ	N (# cm^{-3})	μ (m)	σ	N (# cm^{-3})	μ (m)	σ
Morning	3393	1.0×10^{-8}	1.2	2224	1.35×10^{-8}	1.3	15,532	5.2×10^{-8}	1.7
Afternoon	4132	1.0×10^{-8}	1.2	2232	1.66×10^{-8}	1.2	10,971	4.8×10^{-8}	2
Evening	2066	1.0×10^{-8}	1.2	743	1.55×10^{-8}	1.2	9384.6	5.5×10^{-8}	1.7

Mode-1 (D_p: 5.6–14.3 $\times$ 10^{-9} m); Mode-2 (D_p: 14.3–25 $\times$ 10^{-9} m); Mode-3 (D_p: 25–560 $\times$ 10^{-9} m).

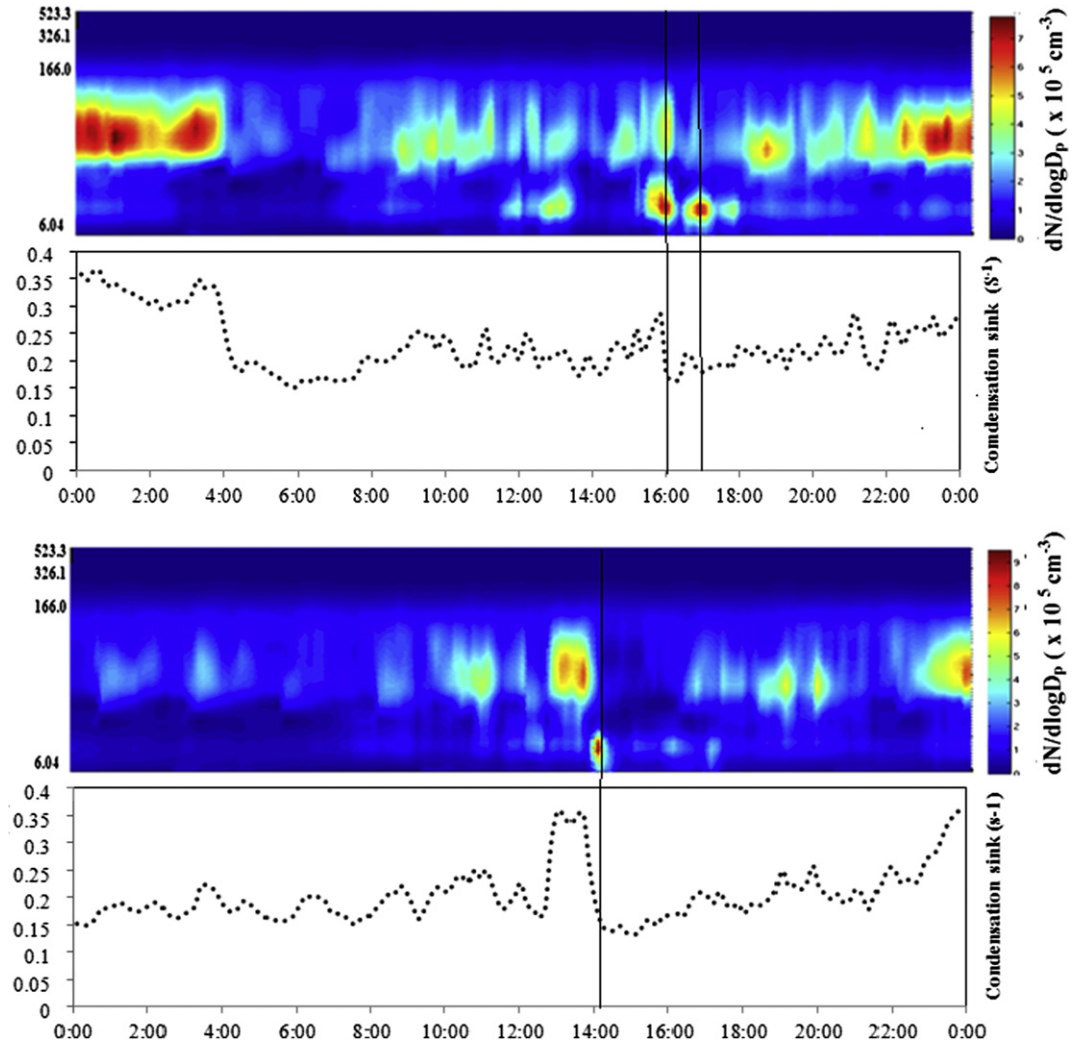

Fig. 6. Particle size distribution plots for 2 different days with nucleation events and condensation sink (CS) profiles for the respective days.

et al., 2008; Asmi et al., 2011; Vakkari et al., 2011). Singapore being highly urban in nature with heavy vehicular and shipping traffic has higher particulate number concentrations and thus higher CS. Similar magnitudes of CS were also reported by Wu et al. (2011) in their study conducted in Beijing. Nucleation events occurred when CS was relatively low, and they were suppressed when CS was high. As can be seen in Fig. 6, there was no significant growth of nucleated particles. It appears that new particles formed in the nucleation mode in this study are significantly different in character compared to those observed at most other locations world-wide (Kulmala et al., 2004). The most likely explanation for this difference is that the nucleated particles at tropical locations undergo micro-scale meteorological phenomena while those at other locations are influenced by regional-scale meteorological phenomena. As discussed in previous sections, these NPF events were observed only when winds blew from the SW direction wherein there were petroleum refineries, acting as point sources of the precursor SO_2 gas. Such nucleation bursts with no significant growth of particles were also reported in a study conducted in the Amazon tropical basin (Rissler et al., 2006). Cheung et al. (2011) also observed nucleation bursts with no growth to large particles in their study conducted at Brisbane. Cheung et al. (2011) reported that these nucleation bursts occurred when winds blew from the NE direction where industries acting as point sources of SO_2 and VOCs were located. These observations are consistent with the finding made in this study. In addition, owing to the convective and turbulent winds in the tropical regions, the freshly nucleated particles seem to have been well-dispersed in the ambient air under the prevailing weather conditions over the sampling site before they could agglomerate and grow, as described earlier. We, therefore, did not calculate the particle growth rates for this study.

The rate of formation, J_5, was calculated using Equation (3) (Dal Maso et al. (2005); Vakkari et al., 2011).

$$J_5 = \frac{dN_{5-25}}{dt} + F_{coag} + F_{growth} \tag{3}$$

The first term dN_{5-25}/dt was obtained from Equation (4)

$$\frac{dN}{dt} = \left[\frac{(C_{peak} - C_{initial})}{(t_{peak} - t_{initial})}\right] \tag{4}$$

where C_{peak} and $C_{initial}$ are the peak number concentrations of the nuclei particles during NPF and the concentration of nuclei mode particles in the ambient atmosphere at the start of NPF. The coagulation loss term F_{coag} was calculated using Equations (5) and (6) (Dal Maso et al., 2005)

$$F_{coag} = CoagS_{nuc} N_{nuc} \quad (5)$$

$$CoagS = \sum K_{ij} N_j \quad (6)$$

where K_{ij} is the coagulation coefficient of particles with diameter i and j. The calculation of coagulation coefficient using the Fuchs (1964) formula is described in detail elsewhere (Seinfeld and Pandis, 1998; Kulmala et al., 2001). As shown in Fig. 6, the new particle formation observed in this study did not have appreciable particle growth. Therefore, F_{growth} representing the losses of particles growing out of the size range was taken to be negligible.

A total of 41 nucleation (NPF) events were observed during the sampling period of two weeks during the SW monsoon period. NPF was identified by a sharp rise in the nucleation mode particle concentrations from the background levels. The rates of formation of new particles (J_5) during the different NPF events during the SW monsoon period along with the meteorological parameters at the time of NPF are presented in Table 4 and their statistical parameters are given in Table 5. Meteorological parameters such as T, RH, and P did not show significant variations (Table 5) while most of the observed nucleation events (26 out of 41) occurred in the temperature range of 29.1 °C to 29.9 °C with the relative humidity in the range of 60%–66%. However as indicated in Table 4, the nucleation rates showed a significant variability (SD = ±10.8 m^3 s^{-1}). We examined the NPF events with the lowest nucleation rate (5.8 m^3 s^{-1}) and the highest nucleation rate (43.5 m^3 s^{-1}) in Table 4. T and SR for both the events were very similar. However, the other parameters such RH and WS were different, indicating the role of meteorological parameters in the new particle formation rates. Nucleation events are sensitive to even slightest changes in meteorological parameters such as temperature and relative humidity. Easter and Peters (1994) reported that small fluctuations in T and RH due to turbulence or mixing processes could lead to considerably higher nucleation rates and turbulence-induced NPF events have been observed (Wehner et al., 2010). In addition to meteorological parameters, there are other factors such as air pollution levels in the atmosphere, especially pre-existing particles with high surface area, that play a key role in the occurrence of NPF events and affect their rate and intensity (McMurry and Friedlander, 1967; Stanier et al., 2004; Kuang et al., 2010). In Singapore's context, although there was no significant change in the meteorological conditions between different NPF events, their intensities varied because of the presence of pre-existing particles in ambient air. The wind directions during nucleation events were mostly from South and South-West directions with a few exceptions. When favorable atmospheric conditions such as high T, high SR, and low pre-existing particle concentrations existed during the afternoon, new particles were formed in the ambient atmosphere.

Table 4
Summary of different nucleation events observed during the SW monsoon period.

Date	Time	Formation rate (J_5) (cm^{-3} s^{-1})	Pressure (hPa)	Air temp (°C)	Relative humidity (%)	Wind speed (m s^{-1})	Wind direction (°)	Incoming radiation (W m^{-2})
30-Jul-08	14:00	33.5	1001	29.2	67	3.5	205	570
31-Jul-08	13:00	11.4	1002	29.6	69	3.3	170	450
31-Jul-08	15:30	29	1000	29.5	63	4.4	170	635
31-Jul-08	17:00	12.7	999	29.6	66	3.8	197	420
1-Aug-08	14:00	39.2	1001	29.7	65	4.7	194	780
1-Aug-08	16:00	10.6	999	30.0	62	3.9	165	535
2-Aug-08	13:00	31.2	1001	30.0	64	1.3	179	815
2-Aug-08	14:15	11.5	1000	30.0	64	1.2	107	220
2-Aug-08	15:45	29.8	1000	29.7	64	3.1	145	680
3-Aug-08	11:30	8.3	1003	28.4	70	3.5	168	650
3-Aug-08	14:15	6.9	1001	29.3	68	2.9	169	470
3-Aug-08	16:00	5.8	1000	29.8	72	5.0	183	675
4-Aug-08	13:30	8.4	1001	29.0	64	4.2	212	750
4-Aug-08	14:45	19.0	1001	29.3	66	2.0	203	730
4-Aug-08	16:30	7.1	1000	29.9	63	0.2	76	565
5-Aug-08	13:15	22.4	1001	30.0	58	3.2	211	665
5-Aug-08	15:00	14.0	1000	29.2	61	4.8	137	690
5-Aug-08	16:30	12.3	999	29.6	61	1.6	204	295
6-Aug-08	10:00	15.8	1002	27.2	75	3.0	158	240
6-Aug-08	15:00	12.5	999	29.4	62	4.8	170	765
6-Aug-08	13:45	25.3	1000	29.2	62	3.7	205	580
7-Aug-08	13:30	14.0	999	29.0	66	4.4	179	280
7-Aug-08	15:00	9.4	998	29.0	66	1.7	139	90
8-Aug-08	10:30	9.9	1001	27.2	69	3.5	217	220
8-Aug-08	13:00	8.2	1000	28.4	71	5.1	203	580
9-Aug-08	11:45	11.8	1002	28.8	66	1.7	227	805
9-Aug-08	15:30	11.8	998	28.0	73	3.3	203	165
10-Aug-08	12:30	7.8	1002	24.4	83	0.2	157	120
10-Aug-08	14:00	6.4	1001	25.0	82	0.2	202	155
10-Aug-08	15:15	7.7	1000	24.8	81	0.2	85	145
11-Aug-08	11:00	26.4	1002	28.8	71	2.5	138	645
11-Aug-08	13:15	35.9	1001	28.8	72	2.0	159	795
11-Aug-08	16:00	43.5	999	29.8	63	2.6	144	665
11-Aug-08	17:00	21.3	999	30.4	59	2.5	204	345
11-Aug-08	18:30	29.2	999	30.1	60	3.3	181	225
13-Aug-08	11:00	29.3	1004	27.1	73	3.6	107	650
13-Aug-08	15:45	19.6	999	29.8	51	2.2	185	785
14-Aug-08	13:30	23.8	1002	29.4	66	4.0	207	915
14-Aug-08	14:00	36.2	1001	29.9	53	2.4	184	855
14-Aug-08	17:00	19.7	999	29.8	63	3.4	169	340
15-Aug-08	13:00	35.4	1001	29.7	56	4.0	206	905

Table 5
Characteristics of nucleation rates and associated meteorological parameters observed during nucleation events.

	Mean	Median	SD	Min	Max
Formation rate J_5 (# cm^{-3} s^{-1})	18.8	14	10.8	5.8	43.5
Surface area (nm^2 cm^{-3})	2.23×10^8	1.98×10^8	8.63×10^7	1.16×10^8	5.25×10^8
Pressure (hPa)	1000	1000	1	998	1004
Air temp (°C)	28.9	29.4	1.4	24.4	30.4
Relative humidity (%)	66.34	65.5	6.4	50.9	83.1
Wind speed (m s^{-1})	3.0	3.3	1.4	0.2	5.1
Wind direction (°)	172.8	179.1	37.0	76.4	227.4
Incoming radiation (W m^{-2})	531.7	580.2	245.7	87.9	912.0

Note: Surface area is the total surface area of pre-existing particles.

Nucleation events can be classified into low (<5000 cm^{-3}), medium (5000–15000 cm^{-3}), and high (>15000 cm^{-3}) intensities according to number of particles formed in the size range (5–25 nm), as proposed by Stanier et al. (2004). It was observed that with an increase in *T* and SR, the intensity of nucleation also increased whereas with an increase in RH and the loading of pre-existing particles, the nucleation intensity decreased (See the Supplementary Section, Fig. S1). These trends are consistent with those reported in earlier studies elsewhere (Jeong et al., 2004; Kulmala et al., 2004). Spearman rank correlation coefficients for the association between these meteorological parameters (*T*, RH, SR) and pre-existing particle surface area and concentrations of NFPs were calculated. RH ($r_s = -0.46$, $p = 0.002$), ambient *T* ($r_s = 0.45$, $p = 0.003$), SR ($r_s = 0.53$, $p = 0.003$), and the particle surface area ($r_s = -0.44$, $p = 0.003$) showed medium to good correlations with concentrations of new particles. The wind speed ($r_s = 0.02$, $p = 0.92$), however, was poorly correlated. Most of the nucleation events observed had limited or no particle growth which was probably due to strong convection, and resulting vertical movement of air which is prevalent in the tropical climate. These turbulent winds tend to disperse newly formed particles before they can be observed to undergo growth.

Nucleation events were rarely observed during the NE monsoon period. As discussed earlier, the prevailing weather conditions such as *T*, RH and SR remained similar to those of the SW monsoon period. One of the reasons for the lack of nucleation events could be due to frequent bush fires occurring in the afternoon time during the NE monsoon period. The bush fires were observed on 21 days during the one month intensive sampling period in January to February. The heavy particle loading in the atmosphere (Fig. 5) most likely suppressed the occurrence of nucleation phenomenon. Such observations pertaining to suppression of NPF events due to biomass burning were reported by Emily et al. (2008) based on their field studies Leeds, UK. The lack of availability of precursor gases such as SO_2 during the NE monsoon could also have contributed to the absence of nucleation events. More systematic field studies in combination with controlled laboratory experiments are needed to gain further insights into NPF events taking place in the urban atmosphere of tropical countries such as Singapore.

4. Conclusions

In this study, particle number concentrations and particle size distributions were investigated in the tropical urban atmosphere of Singapore together with observations of black carbon and SO_2. Proxy H_2SO_4 concentration and CS were estimated during the entire period using the experimental results. Measurements were made during two distinct meteorological periods: the SW monsoon (27 July 2008 through 15 August 2008) and NE monsoon (21 January 2009 to 22 February 2009). Both periods experienced high temperature (*T*) and high incoming solar radiation (SR). During the NE monsoon, winds blew consistently from the NE sector with the sampling site being mainly influenced by local traffic emissions. Local bush fires also occurred during this period and impacted the site sporadically. During the SW monsoon, the measurement site was impacted by the prevailing winds carrying emissions from petrochemical industry and shipping as well as local traffic. New particle formation events were mostly observed during the SW monsoon period, but very rarely appeared during the NE monsoon. The occurrence of NPF was confirmed by rapid increases in PNC (with diameters < 25 nm), combined with a peak in the particle size distribution that was shifted to small particle diameters. The increases in PNC occurred while BC concentrations were low, suggesting that the nanometer-sized particles were not of fossil fuel combustion origin.

The higher frequency of NPF formation during the SW monsoon was most likely due to higher SO_2 concentrations in the air. The high loading of pre-existing particles in the atmosphere during the NE monsoon, caused by local bush fires, may have further suppressed the occurrence of nucleation during this period apart from the lower amount of precursor, SO_2 gas, in the ambient air. NPF events occurred mainly in the afternoon associated with high SO_2 concentrations, higher proxy H_2SO_4 concentrations and low BC concentrations. High *T*, SR, and low relative humidity (RH) conditions were also prevalent during nucleation events. We found that the rate of formation of new particles was dependent on meteorological parameters such as *T*, RH and SR in addition to the concentration of pre-existing particles in the atmosphere. Under the influence of prevailing convective and turbulent winds in the tropical region, the nucleated particles appeared to have been well-dispersed with no significant growth unlike the observations made in mid-latitudes and in boreal forests. Some of the nucleation events occurred at different time points in a day for short durations. One can thus conclude that the nucleation events occurring in the tropical air are relatively short lived with no significant growth of nucleated particles. This study represents the first analysis of NPF in a tropical urban environment. More in-depth field studies are needed to understand the mechanisms behind the nucleation phenomenon in tropical urban environments.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2013.01.049.

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