



# Influence of trans-boundary biomass burning impacted air masses on submicron particle number concentrations and size distributions



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## HIGHLIGHTS

- Aerosol number concentrations and size profiles were investigated.
- Biomass burning impacted air masses were transported above the boundary layer.
- A surge in submicron PNC (1.2–2.5 times) was observed during haze episodes.
- New particle formation was suppressed during haze episodes.
- Particle peak diameters increased during haze episodes.

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## ABSTRACT

Submicron particle number concentration (PNC) and particle size distribution (PSD) in the size range of 5.6–560 nm were investigated in Singapore from 27 June 2009 through 6 September 2009. Slightly hazy conditions lasted in Singapore from 6 to 10 August. Backward air trajectories indicated that the haze was due to the transport of biomass burning impacted air masses originating from wild forest and peat fires in Sumatra, Indonesia. Three distinct peaks in the morning (08:00–10:00), afternoon (13:00–15:00) and evening (16:00–20:00) were observed on a typical normal day. However, during the haze period no distinct morning and afternoon peaks were observed and the PNC ( $39,775 \pm 3741 \text{ cm}^{-3}$ ) increased by 1.5 times when compared to that during non-haze periods ( $26,462 \pm 6017$ ). The morning and afternoon peaks on the normal day were associated with the local rush hour traffic while the afternoon peak was induced by new particle formation (NPF). Diurnal profiles of PNCs and PSDs showed that primary particle peak diameters were large during the haze (60 nm) period when compared to that during the non-haze period (45.3 nm). NPF events observed in the afternoon period on normal days were suppressed during the haze periods due to heavy particle loading in atmosphere caused by biomass burning impacted air masses.

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## 1. Introduction

Airborne particulate matter (PM) has been widely studied in terms of its physical, chemical, and optical characteristics in different parts of the world to understand its influence on environmental and health impacts. Unlike other criteria air pollutants, PM is very heterogeneous and complex in its chemical nature and also comes in various size ranges. Several epidemiological studies

have shown strong associations between PM and increased mortality, morbidity and other respiratory related health effects (e.g. Goldber et al., 2001; Hystad et al., 2013; Nel, 2005; Song et al., 2013; WHO, 2013). These adverse health impacts of PM are particularly pronounced in urban regions due to high population density, intense human activities, proximity to local pollution sources of combustion origin such as on-road vehicles, industries, etc. PM is also known to have climatic impacts on scales ranging from local to global (e.g. Chen and Wu, 2011; Haywood and Boucher, 2000; IPCC, 2007; Mahmud et al., 2010). In addition to local particulate emissions, urban air quality is also affected by regional and inter-hemispheric transport of aerosol particles as has been demonstrated by a number of studies worldwide (e.g. Borge et al., 2007;

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Kindap et al., 2006; Lin et al., 2005; Tchepele et al., 2013). It is therefore desirable to study the significance of local urban particulate emissions and photochemical formation in relation to long-range regional contributions to air pollution in urban environments so that appropriate remedial actions can be taken to improve urban air quality and mitigate its adverse health impacts due to exposure to PM as well as its impacts on climate change (Das and Jayaraman, 2012; Gidhagen et al., 2011).

PM regulatory standards are being constantly upgraded to improve the urban air quality and reduce associated health effects (U.S.EPA, 2013). These regulatory standards are mainly focused on PM mass concentrations. Improved engine technology, development of pollution control devices such as particulate filters, ion catalysts, and alternative energy sources reduced PM mass concentrations emitted from local pollution sources by a large fraction (Goncalves et al., 2009; Sabapathy, 2008). However, particle number concentrations (PNC) still remain a matter of concern as they have not been reduced significantly and in some cases were even increased due to increases in submicron particle emissions from traffic (aerodynamic diameter  $<1 \mu\text{m}$ ) (Betha and Balasubramanian, 2011; Holmen and Ayala, 2002; Kreyling et al., 2003). The submicron particles, especially ultrafine particles ( $d < 0.1 \mu\text{m}$ ; UFPs), make a lower contribution to PM mass than larger size fractions, although they contribute significantly to PNC (Kittelson et al., 2004). It is well-known that UFPs are more harmful to human health compared to larger particles which contribute significantly to particle mass concentration ( $\text{PM}_{2.5}$  and  $\text{PM}_{10}$ ) because of the former's penetrability and high reactive surface area (Chio and Liao, 2008; Nel, 2005). Apart from primary emissions by combustion sources such as vehicles, industries etc., UFPs are also formed in the atmosphere through secondary particle formation via gas-to-particle conversion (Betha et al., 2013; Boy et al., 2008; Kulmala et al., 2004; Shen et al., 2011) and thus play a key role in public health and climatic impacts. Intense field-based investigations are underway to establish PM standards based on number concentrations. The European Union (EU) has recently taken initial steps to set emission regulations for vehicular emissions as  $6 \times 10^{11} \text{ \#}/\text{km}$  (EU, 2012). Owing to the recent developments in particulate emissions and real-time monitoring systems, atmospheric measurements are undertaken to study number and size characteristics of submicron particles in ambient air at various locations all over the world (e.g., Agudelo-Castaneda et al., 2013; Agus et al., 2007; Boogaard et al., 2010; Gomez-Moreno et al., 2011; Hussein et al., 2011; Minoura and Shimo, 2011) to help develop policies to improve urban air quality based on number concentrations as compared to mass concentrations.

Singapore being an urbanized country has several local anthropogenic emission sources which include vehicles, industries, refineries, ships, etc. contributing to ambient particulate pollution. In addition to these local sources, the air quality in Singapore is often affected by episodic "smoke-haze", caused by wild forest and peat fires in the neighboring Indonesian provinces of Sumatra and Kalimantan (Atwood et al., 2013; Velasco and Roth, 2012; Koe et al., 2001; Pavagadhi et al., 2013; Reid et al., 2013). These wild fires occur almost every year during the south-southwest monsoon (S-SW monsoon) season in the region from August to October due to clearing of lands for agricultural purposes. The drier conditions associated with the S-SW monsoon enhance the uncontrolled land clearing activities. The resultant particulate emissions from these wild fires and biomass burning are transported by trans-boundary winds into Singapore and other countries in South East Asia (SEA). Heavy particle loading in the atmosphere during these episodes leads to severe visibility reduction (haze), affecting the local day-to-day activities and public health (Pavagadhi et al., 2013; See et al., 2006). Several studies have been conducted to investigate the

effect of this haze on local air pollution levels and their health impacts in SEA (Koe et al., 2001; Lin et al., 2013; Liu et al., 2013; Pavagadhi et al., 2013; Reid et al., 2013; Salinas et al., 2013). However, particle properties based on number concentrations and PSD profiles during episodes of trans-boundary haze have not been thoroughly investigated to-date. A recent study on the particle number profiles for the tropical urban atmosphere of Singapore has shown clear diurnal variations under two different climatic seasons (Betha et al., 2013), but this study was conducted in the absence of haze.

In the present study, submicron particles were studied in Singapore from 27 June to 6 September, 2009. PNC and PSD were investigated during the occurrence of two trans-boundary haze episodes in the measurement period (6–10 August, 2009). The smoke haze episodes resulting due to wild forest and peat fires in the neighboring country, Indonesia, increased the local atmospheric particle loading. The particle number profiles in Singapore affected by trans-boundary haze due to wild fires in neighboring countries are reported for the first time in this study which are needed for haze-related modeling as well as health risk assessment studies.

## 2. Experimental

### 2.1. Sampling site

Measurements of the number, mass concentrations and size distribution of airborne particles were conducted from 27 June through 6 September 2009 at a tall building with no visible obstructions in the National University of Singapore ( $1^{\circ}18' \text{ N}$ ;  $103^{\circ}46' \text{ E}$ , 67 m a.s.l.). The sampling site (Fig. 1) is 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). A 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 measured by the FMPS every second continuously throughout the sampling period. FMPS was operated at a high flow rate ( $10 \text{ L min}^{-1}$ ) and ambient pressure to minimize particle losses due to diffusion and evaporation of volatile and semivolatile particles as per the manufacturer's recommendation (TSI, 2013). The instrument (FMPS) was connected to a HEPA filter for 5 min every day and 1 h once a week to draw clean air into the instrument to remove any particle deposits on the electrode column. The instrument was then set at zero to ensure the electrometer readings were within

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