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Observation of nucleation mode particle burst and new particle formation events at an urban site in Hong Kong

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HIGHLIGHTS

- Seven NPF events with particle growth were associated with condensable H_2SO_4 vapor.
- Four nucleation bursts were likely caused by incineration in funeral parlor.
- Meteorological conditions play important roles in nucleation mode particle occurrence.

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ABSTRACT

Particle number (PN) concentrations and particle size distributions (PSD) in the size range of 5.5–350 nm were continuously measured from 22 December 2010 to 20 January 2011 at an urban site in Hong Kong when northeastern monsoon prevailed. Apart from the PN peaks appeared in traffic rush hours (i.e. 08:00–09:00 and 17:00–18:00), a distinct peak of PN concentrations in the afternoon (11:00–16:00) was observed during the sampling period. Concurrent measurement data of PSD, ozone (O_3) and proxy sulfuric acid (H_2SO_4) concentrations revealed that the afternoon peaks observed were likely due to new particle formation (NPF) via photochemical reactions. These NPF events were frequently observed under a clean and dry weather in Hong Kong. The occurrence of NPF was closely associated with high solar radiation (SR), low relative humidity (RH) and low condensation sink (CS) in the atmosphere. Besides the NPF events, we also found four nucleation mode particle burst events, typically with increased number concentrations of nucleation mode particles (N_{nuc}) without growth to larger size particles. These burst events were generally accompanied by high-level primary air pollutants, i.e. sulfur dioxide (SO_2), nitrogen oxide (NO_x) and carbon monoxide (CO), low SR and high CS conditions. The very different characteristics of the burst events from those of the NPF events indicated that these nucleation mode particle burst events were not caused by the photochemical reactions, but by the primary emission from the local combustion source(s).

1. Introduction

Air pollution caused by ultrafine particles (UFPs, with a diameter smaller than 100 nm) has become a topic of great interest in terms of air quality impairment, public health and global climate. Typically, UFPs account for about 90% of the total number concentration of particles in the atmosphere (e.g. EPA, 2007; Morawska et al., 2008; González et al., 2011). Atmospheric UFPs comprise two sub-modes, i.e. nucleation mode – (<10 nm) and Aitken mode particles (10–100 nm), and nucleation mode particle has greater deposition fraction in the whole respiratory system compared to larger size particle. In order to better understand the atmospheric UFPs and their impact on atmospheric chemistry, human health, visibility reduction and global climate, it is significantly important to study the sources and the physical/chemical processes of these freshly-formed particles (e.g. Kulmala et al., 2004; Seinfeld and Pandis, 2006; Stölzel et al., 2007; Morawska et al., 2008). Particularly, there is little information about nucleation mode particles in Hong Kong (Yao et al., 2005, 2006; Guo et al., 2012a). Motivated by the above reasons, this study investigated the number concentrations and source origins of UFPs at an urban site of Hong Kong.

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Earlier studies in urban environments claimed that nucleation mode particles are not present in primary exhaust emissions, but are formed through nucleation in the atmosphere after rapid cooling and dilution of emissions when the saturation ratio of gaseous compounds of low volatility (i.e. sulfuric acid) reaches a maximum (Charron and Harrison, 2003; Kittelson et al., 2006; Morawska et al., 2008). Later, some studies found that nucleation mode particles can be either directly emitted from the vehicle exhaust in the particle phase with the composition of elemental carbon, metal species and a fraction of sulfuric acid (H_2SO_4) and organic compounds (unburned oil, etc.) (Kittelson, 1998; Burtscher, 2005; Arnold et al., 2006; Rose et al., 2006; Rodríguez and Cuevas, 2007; Fernández-Camacho et al., 2010), or produced during the dilution and cooling of the exhaust emissions in ambient air from cars (Charron and Harrison, 2003; Yao et al., 2005; Casati et al., 2007), aircrafts (Cheung et al., 2011) ships (Healy et al., 2009; González et al., 2011) and other combustion source like incineration (Karasev et al., 2004; Lipsky and Robinson, 2006; Wright et al., 2007; Géhin et al., 2008; Chuang et al., 2011; Manoukian et al., 2013). All the studies indicated that the nucleation mode particles emitted from primary sources are often accompanied by high-level co-pollutants i.e. SO_2, NO_x and/or CO, typical of direct source emissions.

Apart from direct sources, nucleation mode particles are also newly formed in the atmosphere through gas-to-particle conversion. This phenomenon is a relatively complex photochemical process commonly defined as atmospheric nucleation or new particle formation (NPF), which usually shows subsequent growth of particle size (e.g. Holmes, 2007; Guo et al., 2008; Kulmala and Kerminen, 2008; Mikkonen et al., 2011a). A number of NPF studies in different environmental settings around the world demonstrate that NPF is typically favored under the conditions of low pre-existing aerosols, as their surface area serves as a condensation and coagulation sink of ambient vapors and small particles (Kulmala et al., 2004; Hamed et al., 2007; Neitola et al., 2011; Shen et al., 2011; Vakkari et al., 2011). As such, the elevated background concentration of aerosols in polluted locations appears to suppress the NPF. Nevertheless, the NPF was still observed in some polluted environments and most of the studies have related the NPF to the concentration of vapor-phase H_2SO_4 which is mainly generated by the oxidation of SO_2 by the hydroxyl (•OH) radical during daytime (e.g. Weber et al., 1997, 1999; Jeong et al., 2004; Sihto et al., 2006; Riipinen et al., 2007; Kuang et al., 2008; Petäjä et al., 2009; Mikkonen et al., 2011b).

In Hong Kong about 97% of SO_2 emissions are from local marine sources (50%) and power plants (47%) (http://www.epd.gov.hk/epd/tc_chi/environmentinhk/air/data/emission_inve.html). In addition, under the prevailing north/northeast synoptic winds in winter, the regional polluted air enriched with SO_2 from inland Pearl River Delta (PRD) often reaches Hong Kong. Hence, we presented one-month data of particle number (PN) concentrations and particle size distributions (PSD) with a size range of 5.5–350 nm in winter (from December 2010 to January 2011) to investigate the intensity and frequency of nucleation events at an urban site. The characteristics of nucleation mode particle burst and the NPF events were discussed in details, by looking into the simultaneously monitored trace gases (i.e. O_3, SO_2, NO–NO_2–NO_x and CO) and meteorological parameters.

2. Methodology

2.1. Site description

The sampling site, situated on the rooftop of a building in the campus of Hong Kong Polytechnic University at Hung Hom, Hong Kong (HKPolyU site; 22.3° N, 114.177° E, about 40 m a.s.l), is located near main roads and the cross-harbor tunnel (CHT) (Fig. 1). The daily average number of vehicles passing through the CHT was about 122,000 in December 2010–January 2011, of which, about 30% were diesel-fueled vehicles, 60% gasoline-fueled vehicles, and 10% liquefied petroleum gas (LPG)-fueled taxies (HKTD, 2010, 2011). The traffic peaks appear at 08:00–09:00 in the morning and 17:00–18:00 in the afternoon and evening (Xia and Shao, 2005; Cheng et al., 2012). To the northeast of the sampling site, there are two funeral parlors with a distance of ~500 m. To the southwest and southeast is the Victoria Harbour. Hence, the study area could be affected by traffic emissions, incineration from the funeral parlor and marine vessel emissions (Fig. 1).

2.2. Measurement techniques

A Scanning Mobility Particle Sizer (SMPS, model 5.400, GRIMM, Germany), coupled with a butanol-based Condensation Particle Counter (CPC, model 5.400, GRIMM, Germany), was employed to continuously measure the PN concentrations and PSDs in the range of 5.5–350 nm at a 4-min scan interval during the sampling period. To minimize the particle losses caused by electrostatic attraction and diffusion deposition, a 1.5 m long conductive tube with an inner diameter of 0.8 cm and a temperature-controlled room were utilized for the SMPS + CPC sampling system. Detailed description of the SMPS + CPC system and its calibration was given in Wang et al. (2012). In addition, the measurement data of SMPS + CPC for identified nucleation mode particle burst events were checked to exclude any data caused by instrumental artifacts, because the SMPS + CPC instrument could malfunction by reading artifact signals in the nucleation mode due to air leak in the system (e.g. Hogrefe et al., 2006) or extremely high particle number concentrations introduced into the system (e.g. Joshi et al., 2012). Firstly,

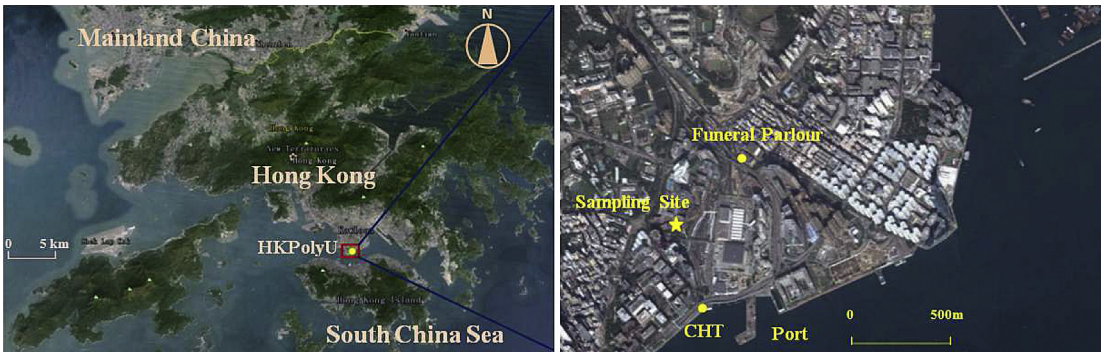

Fig. 1. The sampling site and the surrounding environment.