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Temporal distribution and other characteristics of new particle formation events in an urban environment☆

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ABSTRACT

Studying the characteristics of new particle formation (NPF) is important as it is generally recognized as a major contributor to particle pollution in urban environments. We investigated NPF events that occurred during a 1-year period in the urban environment of Brisbane, Australia, using a neutral cluster and air ion spectrometer (NAIS) which is able to monitor both neutral and charged particles and clusters down to a size of 0.8 nm. NPF events occurred on 41% of days, with the occurrence rate of 7% greater in the summer than in the winter. We derived the first diurnal event distribution of NPF events anywhere in the world and showed that the most probable starting time of an NPF event was near 08:30 a.m., being about an hour earlier in the winter than in the summer. During NPF days, 10% of particles were charged. The mean neutral and charged particle concentrations on NPF days were, respectively, 49% and 14% higher than those on non-event days. The mean formation rate of 2–3 nm particles during an NPF event was 20.8 cm^{-3} s^{-1}. The formation rate of negatively charged particles was about 10% higher than that of positively charged particles. The mean particle growth rate in the size range up to 20 nm was 6.2 nm h^{-1}. These results are compared and contrasted with corresponding values that have been derived with the scanning mobility particle sizer (SMPS) at the same location and with values that have been reported with the NAIS at other locations around the world. This is the first comprehensive study of the characteristics of NPF events over a significantly long period in Australia.

1. Introduction

Secondary particles are formed by the homogeneous nucleation of gaseous precursors such as sulphuric acid, ammonia and volatile organics, in the atmosphere (Kulmala et al., 2013). This process is also known as new particle formation (NPF) and has been observed in many different environments including urban, industrial, agricultural and coastal sites, as well as in boreal forests and the polar regions (Kulmala et al., 2004). The large majority of these observations were made in the northern hemisphere, mostly in Europe.

In the atmosphere, gas molecules cluster together and remain stable until they reach a size of about 1.6 nm. These are known as clusters (Hirsikko et al., 2011). Beyond this size, if the gaseous saturation remains high enough, they go on to form particles. NPF typically occurs around mid-day with high intensity of solar radiation, high concentration of gaseous precursors and low concentration of pre-existing aerosols (Birmili and Wiedensohler, 2000). The role of ions in this process has also been proposed (Iida et al., 2006; Yu and Turco, 2000). Ions attach to neutral gas molecules in the air to form charged molecular clusters smaller than 1.6 nm in size. These “cluster ions” attach to aerosol particles in the air, producing charged particles. Charged particles in the size range 1.6–7.5 nm are known as intermediate ions and are used as an indicator of particle formation in the atmosphere as their concentration shows a sharp increase during NPF events.

There have been several studies of NPF in Brisbane. For example, Guo et al. (2008) reported NPF events on 7 out of 20 days (35%), while Cheung et al. (2011) identified NPF events on 65 out of 252 days (26%). In contrast, Salimi et al. (2017) reported 219 NPF events out of 285 days of measurements at 25 sites across Brisbane. This occurrence rate of 77% is significantly higher than any of the values found previously in Brisbane and at any other location in the world. These three studies were carried out using a scanning mobility particle sizer (SMPS) with a lower size limit of about 10 nm.

In the present study, we collected data of charged and uncharged particle concentrations in the urban environment of

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Brisbane using a Neutral cluster and Air Ion Spectrometer (NAIS) over a one year time period. The NAIS is designed to measure particles down to a size of 0.8 nm and does not have the limitations of the SMPS in identifying NPF events. We have also previously used a NAIS in Brisbane to investigate various characteristics of NPF events. Crilley et al. (2014) monitored particle number concentration (PNC) and the chemical composition of aerosols using a NAIS and a time-of-flight aerosol mass spectrometer (TOF-AMS) and showed that NPF events were observed on 20 out of 36 days (55%). Airborne PNC, charged particles, sulphate and ammonium concentrations were highly correlated on NPF days. Jayaratne et al. (2015b) showed that cluster ion concentrations were suppressed by high PNCs and NPF events were more likely to form when the air was cleaner. Jayaratne et al. (2016b) investigated the charging state of aerosols during NPF events and showed that NPF occurred on 45% of days observed. The coefficient of unipolarity (positive/negative ion concentration) was 1.37 for cluster ions and 1.17 for charged particles. The positive cluster ion concentration was 40% higher than negative while the positive charged particle concentration was 20% higher than negative. The cluster ion concentration was higher during the night than in the daytime, while the charged particle concentration was higher during the daytime than in the night. There was a positive correlation between PNC and charged particle concentration and a negative correlation between PNC and cluster ion concentration. The percentage of particles carrying a charge was 10–15% during the night and 5–10% during the day.

In this study, we derived the characteristics of NPF such as the particle formation rates and growth rates using PNC down to the smallest particle sizes for the first time in Brisbane. In addition, we estimated the short and long term temporal variations of the occurrences of NPF and derived a diurnal distribution plot of NPF events for the very first time anywhere in the world.

2. Methods

2.1. The study area

The measurements were conducted at the Garden Point campus of the Queensland University of Technology in Brisbane, Australia, which is a typical urban environment, over one calendar year. Monitoring was carried out through the window of a building at a height of approximately 10 m above the ground, about 100 m away from a busy highway. The dominant anthropogenic sources at this site are motor vehicle exhaust emissions. Other anthropogenic sources affecting the site are emissions from the Brisbane port and two oil refineries located about 20 km to the north-east of the city.

2.2. Instrumentation

The NAIS is manufactured by Airel Ltd, Estonia (Manninen et al., 2016). It is designed to detect neutral and charged cluster and particles in the electrical mobility range from 3.2 to 0.0013 $cm^2\ V^{-1}\ s^{-1}$. This corresponds to a particle size range from 0.8 to 42 nm. It has a high-resolution time down to 1 s. The instrument cycles between the ion mode, where it measures naturally charged particles, and the particle mode in which it measures both charged and uncharged particles. Two parallel mobility analysers enable both positive and negative particles to be monitored simultaneously.

Manninen et al. (2011) and Manninen et al. (2016) have recently pointed out that the NAIS has some difficulty in differentiating between charged particles smaller than 2.0 nm and corona ions generated by the charger in the instrument. Therefore, as recommended by these workers, we have limited the smallest particle that can be detected by the NAIS to 2.0 nm. Since the NAIS does not detect any particles larger than 42 nm, all PNC values quoted in this paper refer to the size range 2–42 nm.

A scanning mobility particle sizer (SMPS), including a TSI 3071 differential mobility analyser and a TSI 3782 condensation particle counter, was used to measure the particle size distribution in the range 9–415 nm. The data obtained with the SMPS were used to calculate the condensation sinks.

2.3. Data analysis

2.3.1. Classification of NPF events

NPF events were identified using the rate of change of total particle concentration, dN/dt, where N is the number of particles in the size range 2.0–10.0 nm and using the classification scheme developed by Zhang et al. (2004). Events with N > 10,000 cm^{-3} for at least 1 h and dN/dt > 10,000 $cm^{-3}\ h^{-1}$ were classified as NPF events. These events generally exhibit a “banana” shape in the contour plot of PNC. Days where the above criteria are not fulfilled were classified as “non-event” days.

2.3.2. Starting time of NPF events

In the absence of NPF, the PNC in the intermediate size range 2.0–7.5 nm is very small. When an NPF event begins, this PNC shows a sharp increase. This increase may be used to identify the starting time of an NPF event (Leino et al., 2016). In this study, we used the time of first occurrence of dN/dt > 10,000 $cm^{-3}\ h^{-1}$, where N is the number of particles in the size range 2.0–10.0 nm, as the starting time of an event.

2.3.3. Calculation of condensation sink (CS) and coagulation sink (CoagS)

The condensation sink determines the condensation rate of the vapour onto the aerosol particles. This parameter is controlled by the diffusion properties of condensing vapour and the surface area of aerosol particles.

The CS of particles is defined as

$$CS = 2\,\pi\,D\sum_{\boldsymbol{i}}\beta_m(d_{\boldsymbol{p},\boldsymbol{i}})d_{\boldsymbol{p},\boldsymbol{i}}N_i \tag{1}$$

where D is the diffusion coefficient of the condensing vapour and β_m is the transition correction factor for mass flux (Dal Maso et al., 2002; Lehtinen et al., 2003; Dal Maso et al., 2005; Salma et al., 2011; Kulmala et al., 2012). In this expression, $d_{p,i}$ and N_i are the diameter and the number concentration of particles in the size bin i, respectively. The unit of CS is per second. In this study, we calculated the CS from the number concentration of particles reported by the SMPS in 107 size bins in the range 9–415 nm and by the NAIS in 7 size bins in the range 2–9 nm.

Several studies have shown that sulphuric acid was the main condensing vapour in the urban atmosphere (Stanier et al., 2004; Zhang et al., 2004). Studies in Brisbane have confirmed this result (Crilley et al., 2014; Salimi et al., 2014). We calculated the diffusion coefficient of sulphuric acid using the expression given by Jeong (2009):

$$D = \left(5.0032\times10^{-6}\right) + \left(1.04\times10^{-8}T\right) + \left(1.64\times10^{-11}T^2\right) - \left(1.566\times10^{-14}T^3\right) \tag{2}$$

where D has the units of square metres per second and temperature T is in Kelvin. Here, we used T = 300 K which is a good estimate of the average daytime temperature in Brisbane. The value of D