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Impact of particle formation on atmospheric ions and particle number concentrations in an urban environment

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ABSTRACT

A measurement campaign was conducted from 3 to 19 December 2012 at an urban site of Brisbane, Australia. Size distribution of ions and particle number concentrations were measured to investigate the influence of particle formation and biomass burning on atmospheric ion and particle concentrations. Overall ion and particle number concentrations during the measurement period were found to be (-1.2×10^3 cm^{-3} | $+1.6 \times 10^3$ cm^{-3}) and 4.4×10^3, respectively. The results of correlation analysis between concentrations of ions and nitrogen oxides indicated that positive and negative ions originated from similar sources, and that vehicle exhaust emissions had a more significant influence on intermediate/large ions, while cluster ions rapidly attached to larger particles once emitted into the atmosphere. Diurnal variations in ion concentration suggested the enrichment of intermediate and large ions on new particle formation event days, indicating that they were involved in the particle formation processes. Elevated total ions, particularly larger ions, and particle number concentrations were found during biomass burning episodes. This could be due to the attachment of cluster ions onto accumulation mode particles or production of charged particles from biomass burning, which were in turn transported to the measurement site. The results of this work enhance scientific understanding of the sources of atmospheric ions in an urban environment, as well as their interactions with particles during particle formation processes.

1. Introduction

Atmospheric ions are mainly formed by the ionization of air molecules by cosmic rays from space, or alpha radiation from natural radioactive materials such as Rn-222 in the ground and its airborne progeny (Reiter, 1992). In addition, an increase in cluster ion concentration has also been observed during nucleation events in boreal environments (Vana et al., 2006). Moreover, ions are known to be emitted from combustion processes, hot surfaces and flames (Maricq, 2006; Fialkov, 1997; Peineke and Schmidt-Ott, 2008), as well as during the break-up of water droplets (Laakso et al., 2007). In urban environments, vehicle emissions are the major source of many pollutants including ions and charged particles. Cluster ion concentration (<2 nm) measured near roadside and motorway sites has been found to be several times higher than that in the urban background (Jayaratne et al., 2010).

Previous studies of ion measurements have focused on contribution of ions (i.e. ions induced nucleation, IIN) to the particle formation process in rural areas (e.g., Laakso et al., 2004; Hirsikko et al., 2007), whereas limited studies have been conducted in urban environments (Hirsikko et al., 2011). In an urban setting, particle sources are more complex than those in rural areas due to the mixing of different combustion emissions, such as those from transportation (e.g. motor vehicles, ships and aircraft), industrial sources, power plants, and occasionally biomass burning and eolian dust particles

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from rural areas (Morawska et al., 2008; González and Rodríguez, 2013). Different sources have their own pollution characteristics which interact with each other, thereby making it difficult to interpret the resulting data in relation to the physico-chemical properties and formation mechanisms. Elevated particle number concentration (PNC) associated with new particle formation (NPF) events has been frequently observed in Brisbane during the warm season (Cheung et al., 2011). Kalivitis et al. (2012) found a negative correlation between cluster ion and black carbon concentrations where the loss of cluster ions was due to ion attachment on the accumulation particles associated to the black carbon source in the study area. Although the characterization of atmospheric particles associated with NPF and biomass burning has been studied, very few reports were made pertaining to ions and charged particles. The study of atmospheric ions is vital, not only due to their potential impacts on human health, but also their involvement in atmospheric particle formation processes (Fews et al., 1999; Henshaw, 2002; Hirsikko et al., 2007).

The aim of this study was to investigate the influence of NPF and biomass burning on the variation of atmospheric ion and particle number concentrations in an urban environment. In this paper, we present the size distribution of ions and PNC measured in an urban area of Brisbane, Australia. New particle formation and the transport of biomass burning emissions from upwind of the site were both observed in the study area where ambient levels of ion and aerosol particles were elevated. To our knowledge, this is the first report on the variations of ion size distribution under the influence of biomass burning in an urban environment. The results enhance our understanding of the sources of atmospheric ions and their interactions with particles during the particle formation process.

2. Methodology

2.1. Study design

The measurements were conducted at the International Laboratory for Air Quality and Health (ILAQH), Queensland University of Technology (QUT) (see Fig. 1 for map). The monitoring site was about 10 m a.g.l. on the top floor of a QUT campus building, which lies to the south of the city center, with a major highway (the Pacific Motorway) carrying about 120,000 motor vehicles per day situated about 100 m away along the southwest of the site. The other major sources of air pollutants in Brisbane include the airport, oil refinery and Port of Brisbane located about 10 km to the northeast of the campus, and a gas-fired power plant located about 40 km to the southwest of the sampling site. Therefore, the pollution associated with the northeasterly winds could be mainly attributed to industrial and aircraft/ship emissions, while that associated with southerly to northwesterly winds was due mostly to local traffic exhaust emissions in addition to the plumes of power plant.

Monitoring was conducted from 3 to 19 December 2012, when nucleation events were most likely to occur (Cheung et al, 2011). Furthermore, controlled burning generally continued around Brisbane at this time of the year and we wished to investigate the effect of biomass burning emissions on the ion concentrations. In the context, the results of this investigation should be attributed generally to the characteristics of ions and aerosols for biomass burning season in Brisbane. During the sampling period, prevailing synoptic winds in the Brisbane area were from the northeast, except from 10 to 15 December, when southeasterly winds dominated. During the study period, the mean temperature was observed to be 27 °C, with minimum and maximum temperatures of 18.9 °C and 41.9 °C, respectively. A light breeze (~1–3 m s^{-1}) was generally observed, with light precipitations on 8th (0.6 mm), 10th (0.6 mm), 11th (5.4 mm) and 19th (1 mm) of December.

2.2. Measurement techniques

The size distributions of ions and charged and neutral particles were measured by a Neutral cluster and Air Ion Spectrometer (NAIS, Airel Ltd.) at the QUT monitoring site. The detection size range of the instrument was from 0.8 to 42 nm for both ions and particles. The NAIS consisted of two identical electrical mobility analyzer columns, one for each polarity, so that ions of both signs could be monitored simultaneously. The aerosols were size-classified by the mobility analyzers and measured with an array of 21 electrometers in each column. The air sample was drawn into the NAIS from outside the building through a 2.54 cm (inner diameter) conductive rubber tube with a length of 1100 mm, at a sampling flow rate of 60 Lpm (30 Lpm in each column). The instrument is designed to sample in a cycle and the sampling duration was set to 2 min per sample. PNC was continuously measured by a Condensation Particle Counter (CPC, TSI 3787) which counted total number of particles from the size range of ≥ 5 nm. The sample was drawn through a 6.35 mm (inner diameter) conductive rubber tube and the data time resolution was 1 min. Sulfur dioxide concentration (SO_2) was measured by a pulsed UV fluorescence analyzer (Ecotech 9850, with a detection limit of 0.5 ppbv and accuracy of $\pm 1\%$ of instrument reading) with a sampling interval of 5 min. Span and zero calibrations were conducted before and after the study.

In addition, carbon monoxide (CO), nitrogen dioxide (NO_2) and mass concentration of $PM_{2.5}$ and PM_{10}, as well as meteorological parameters (including wind speed/direction, temperature, relative humidity, and global solar radiation) measured by Queensland Department of the Environment and Heritage Protection (EHP) in the city of Brisbane were used to assist with the data analysis. A detailed description of the instruments used for the acquisition of meteorological data is available at the EHP website (http://www.ehp.qld.gov.au/air/pollution/pollutants/index.html).

2.3. Data processing and analysis

During the sampling period, the NAIS data was found to be affected by emissions from cleaning equipment operated at 06:00 LT on campus during weekdays, therefore the corresponding data (~30–60 min) was removed from the database. Spurious events also resulted in occasional data spikes on the NAIS. In order to eliminate these, the upper limit of the ion concentrations was set to 10^5 cm^{-3}. Particle diffusional loss through the sampling tube in the NAIS was calculated as ~3% at 2 nm ion size, thus no diffusional loss corrections were applied to ion concentrations. The ion data were then classified into