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Suppression of Cluster Ions during Rapidly Increasing Particle Number Concentration Events in the Environment

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ABSTRACT

We show that the cluster ion concentration (CIC) in the atmosphere is significantly suppressed during events that involve rapid increases in particle number concentration (PNC). Using a neutral cluster and air ion spectrometer, we investigated changes in CIC during three types of particle enhancement processes – new particle formation, a bushfire episode and an intense pyrotechnic display. In all three cases, the total CIC decreased with increasing PNC, with the rate of decrease being greater for negative CIC than positive. We attribute this to the greater mobility, and hence the higher attachment coefficient, of negative ions over positive ions in the air. During the pyrotechnic display, the rapid increase in PNC was sufficient to reduce the CIC of both polarities to zero. At the height of the display, the negative CIC stayed at zero for a full 10 min. Although the PNCs were not significantly different, the CIC during new particle formation did not decrease as much as during the bushfire episode and the pyrotechnic display. We suggest that the rate of increase of PNC, together with particle size, also play important roles in suppressing CIC in the atmosphere.

Keywords: Aerosol particle; Charged particle; Particle formation; Bushfire.

ABBREVIATIONS

COU	Coefficient of Unipolarity
NPF	New Particle Formation
PNC	Particle Number Concentration
CIC	Cluster Ion Concentration
NAIS	Neutral cluster and Air Ion Spectrometer

INTRODUCTION

Air molecules are continuously ionized by cosmic rays and radioactivity from radon and its daughter products in the environment. The resulting electrically charged molecules are quickly clustered by other molecules, principally water vapour, and form cluster ions, also known as 'small ions' (Hirsikko *et al.*, 2011). Cluster ions are thermodynamically stable until they grow to a size of about 1.6 nm (Horrak *et al.*, 2000). They are found in concentrations of 200–2500 cm^{-3} in unpolluted environments, but their concentrations may be greatly elevated in the presence of anthropogenic sources of ions such as overhead power lines (Buckley *et al.*, 2008; Jayaratne *et al.*, 2008) and motor vehicles (Jayaratne *et al.*, 2010). Cluster ions have a limited lifetime as they are soon lost by recombination or attachment to aerosol particles in the air. Particles that are charged in this manner are generally larger than 1.6 nm and also known as 'large ions' (Hirsikko *et al.*, 2011). Thus, the cluster ion concentration (CIC) is highly dependent on the particle number concentration (PNC) in the air. As the production rate of cluster ions at a given location is fairly constant in time, the CIC is determined by the loss rate which is mainly due to two processes – recombination with other clusters and attachment to particles. In polluted urban areas where the PNC is relatively high, the attachment process dominates over recombination of small ions and, so, the CIC is mainly controlled by the PNC (Retalis *et al.*, 1991). If the PNC is stable, the CIC will attain a balance value, and this is generally what is observed in the natural environment. While the CIC often exceeds 10^3 cm^{-3} in clean environments (Hirsikko *et al.*, 2011), it may be as low as 200–400 cm^{-3} in polluted urban environments (Hirsikko, 2007; Retalis *et al.*, 2009; Ling *et al.*, 2010). Under stable conditions, atmospheric particles attain an equilibrium charge distribution due to bipolar diffusion charging. The fraction of particles that are charged increases with particle size, typical equilibrium values being about 9% at 10 nm and 35% at 40 nm (Wiedensohler, 1988). Thus, if the PNC and the particle size distribution do not vary in time, the CIC will remain steady. However, if the PNC increases rapidly, charge equilibrium is not achieved and, as a result, the particles

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will be 'under-charged' and the CIC will decrease sharply.

Rapid increases in PNC are commonly observed during new particle formation (NPF) events in the urban environment (Woo *et al.*, 2001). They involve the rapid formation of secondary particles by the homogeneous nucleation of gaseous precursors such as sulfuric acid, ammonia and organics, mainly from motor vehicle emissions and industrial sources (Kulmala, 2003). During such events, the PNC in the environment can increase from a few thousand cm^{-3} to over 1×10^5 cm^{-3} within time periods of 1–2 h (Woo *et al.*, 2001). Most of the particles formed will be in the nanoparticle size range, that is, smaller than 50 nm (Morawska *et al.*, 2009). Previous studies in Brisbane have shown that NPF is a regular occurrence (Cheung *et al.*, 2011).

Very sharp localized increase in PNC within a very short time can also occur during a pyrotechnic display (Wehner *et al.*, 2001) and provides an ideal situation to investigate any accompanying decrease in CIC. Extreme PNCs are also observed due to bushfires which comprise a major source of particle pollution in the atmosphere. Controlled bush burning is periodically carried out around the city of Brisbane to reduce the amount of ground vegetation, decrease the risk of bushfires that could cause greater damage and assist in maintaining biodiversity. The resultant smoke is sometimes observed to extend to and envelop the central city area.

In this study, we monitored cluster ions and charged particle concentrations and neutral PNC at an urban location using a neutral cluster and air ion spectrometer (NAIS) during several NPF events, a controlled bush burning episode that continued over three days and affected the monitoring site, and an intense pyrotechnic display that took place around the monitoring site and lasted for a full 25 min. Our main aim was to investigate the effect of PNC on the ambient CIC. Although there are many studies of the effect of PNC on CIC (Retalis *et al.*, 2009; Hirsikko *et al.*, 2011; Ling *et al.*, 2013), there are few reports, if any, of the interaction rate of cluster ions and particles during rapid PNC variations that tend to occur frequently in the environment.

METHODS

Instrumentation

The instrument used in this study was the Neutral cluster and Air Ion Spectrometer (NAIS), manufactured by Airel Ltd, Estonia (Mirme *et al.*, 2007). The instrument contains two electrical mobility analysers, one for each polarity, preceded by two pairs of unipolar corona wire diffusion chargers and electrostatic post-filters to provide controlled charging, enabling it to distinguish neutral particles from charged ions. In this way, the NAIS can measure positive and negative cluster ions and charged particle concentrations simultaneously as well as neutral clusters and particles in the mobility range 3.16 to 0.001 $cm^2/V's$, which corresponds to a mobility size range of 0.8 to 42 nm. The upper size detection limit of the NAIS effectively restricts it to the nanoparticle size range, which has somewhat arbitrarily been defined as smaller than 50 nm (Morawska *et al.*, 2009). Therefore, all PNCs in this paper refer to nanoparticles only. In this study, we set the NAIS to operate in a cycle of 2.5 min including ion and neutral particle sampling periods of 1.0 min each, the remaining 0.5 min being an offset period which is required to neutralize and relax the electrodes. For a good description of the basic operational principle of the NAIS, the reader is referred to Manninen *et al.* (2009).

Sampling Techniques

The NAIS was located in a laboratory on the top floor of a six-floor building at the Gardens Point Campus of the Queensland University of Technology (QUT) in Brisbane, Australia. The site bordered the main city centre and a large city park and was situated about 100 m away from a busy freeway. The air was sampled from outside a window through a conductive rubber tube of length 1.1 m and diameter 17 mm. A steel mesh was attached to the inlet of the tube to prevent windblown debris and insects, such as flies and mosquitoes, from being sucked into the instrument in the air flow. The instrument was operated by the software provided on an on-line computer which also stored the data acquired in real time.

Study Design

The NAIS was used to sample ambient air intermittently over the calendar year 2012. The ambient data used in this paper is based on the measurements carried out over the month of April, 2012. This period was chosen as it included 30 continuous days of data with no instrumentation problems or interruptions. An intense pyrotechnic (fireworks) display took place as part of the Annual Riverfire Festival in Brisbane on the evening of 29th September 2012. The display encompassed a large area along a 1 km stretch of both banks of the Brisbane River, with pyrotechnics fired into the air from the tops of several tall buildings, some of which were located immediately next to the building from where the monitoring was being conducted. During the period 16–19 December, 2012, controlled bush burning operations were conducted some 100–200 km to the north of Brisbane. Northerly winds that developed on the 17th and 18th December carried high concentrations of biomass burning pollutants to the monitoring site.

For purposes of comparison, we also used PNC and CIC data obtained at a rural site located well away from human activities where the air was clean and near the curb of a busy motorway where the PNC was relatively high. These two studies are described in two earlier publications (Ling *et al.*, 2013; Jayaratne *et al.*, 2014).

Data Analysis

The NAIS was operated continuously during each period of measurement. One minute average readings of ion and PNC were obtained in each 2.5 min cycle, giving 24 readings of each parameter every hour. Differences between means of samples were estimated statistically using the Student's t-test. Linear regression analyses were used to investigate significant increases or decreases of parameters. All significant differences in both the t-tests and the linear regression analyses were estimated at a confidence level of 95%.

NPF events were identified based on the rate of increase of PNC in the size range 1.6–10 nm, dn/dt. Events with dn/dt > 15,000 cm^{-3}/h were classified as PF events in accordance with the method employed by Zhang *et al.* (2004). Days with no such events were classified as non-event days.

The coefficient of unipolarity (COU) is defined as the ratio of the positive to the negative ion concentrations (Gefter, 2002; Kolarž *et al.*, 2009) and the COUs for clusters and nanoparticles were calculated separately.

RESULTS AND DISCUSSION

Days with No Particle Formation Events

NPF events were observed on 12 of the 30 days investigated. The criterion used to identify an NPF event is described in the previous section. Generally, on days with no NPF events, PNC and charged particle concentrations showed good correlation to each other while the total CIC showed an anti-correlation to each of the two parameters separately. This is explicable as cluster ions are rapidly scavenged by particles in the air. Fig. 1 shows the PNC, charged particle concentration and total CIC on 24 April which was a typical day with no particle formation events. The curves indicate the one-hour running averages. The PNC and charged particle concentration were minimum at night and they both increased during the day when there was more human activity; the daytime values being most influenced by motor vehicle emissions. The morning peaks, between 6:00 and 8:00 h coincided with the morning rush hour traffic. The absence of a late afternoon rush hour peak is a consequence of greater atmospheric turbulence that serves to disperse particles vertically. The curves in Fig. 1 show that the PNC and total charged particle concentration did not exceed 5×10^4 cm^{-3} and 5×10^3 cm^{-3}, respectively, at any time during the 24 hour period. Conversely, the total CIC was a maximum during the night (800–1000 cm^{-3}) and decreased significantly (to 500–700 cm^{-3}) during the day. Cluster ions and charged particles coexisted in the air at all times, with a small excess of positive over negative ion concentrations, reflected by consistently larger than unity COUs. The mean and standard deviations of the cluster ion and charged particle COUs were 1.4 ± 0.4 and 1.2 ± 0.2, respectively.

Days with Particle Formation events

Fig. 2 shows a similar set of Results obtained on 7 April, which was characterised by two particle burst events. There was a very sharp increase in PNC between 9:00 and 10:00 and a less intense event between 15:00 and 17:00. The temporal variation of the particle size distributions on this day, observed by the NAIS, showed the typical 'banana shape' associated with NPF during the morning event (Fig. 3). However, although there was an increase in PNC in the afternoon, this was not a sharp increase and did not satisfy the criteria identifying an NPF event as specified in section 2.4. During the morning event, the PNC increased by almost an order of magnitude - from about 2.5×10^4 cm^{-3} to over 2.0×10^5 cm^{-3} in just one hour. Fig. 2(b) shows that the corresponding total charged particle concentration increased from about 2.5×10^3 cm^{-3} to 1.0×10^4 cm^{-3}. Fig. 2(c) shows that, during this period, the average total CIC halved – from 850 cm^{-3} to 425 cm^{-3}. This is a dramatic decrease in CIC and it occurs due to the rapid increase in PNC. Note also that, while the PNC increased by an order of magnitude, the charged particle concentration only increased by a factor of about 4. This indicates that the particles were under-charged. The production rate of cluster ions was not sufficiently fast to keep pace with the rapidly increasing PNC. This phenomenon was observed during all of the 12 NPF events, especially when the PNC increased rapidly. In general, both the positive and negative CICs were suppressed during NPF events with the cluster ion COU greater than unity at all times.

Bushfire Event

Fig. 4 shows the PNC, charged particle concentration and total CIC on 18 December when the air at the monitoring site was influenced by pollutants from the biomass burning event. The wind changed from east to north at about 8:00, bringing smoke to the monitoring site. The PNC began to increase at about 9:00 and reached over 1.4×10^5 cm^{-3} within the next five hours. During this time period, the atmospheric optical visibility remained low with a particle back scattering coefficient larger than 90 Mm^{-1} and an average wind speed of 1.6 m/s. The absence of a 'banana shape' in the temporal variation of the particle size distribution indicated that this was not an NPF event and that the particles were entrained to the site from elsewhere. The smoke lasted in the environment until about 14:00 when an easterly change in the wind, coupled with an increase in wind speed to 3.0 m/s, led to its rapid dispersion. During the five hour period influenced by smoke, the total CIC remained below 150 cm^{-3}, which was significantly less than the previous night-time values of 300–500 cm^{-3} (Fig. 4(c)). As the smoke entrained the monitoring site, the PNC increased by a factor of 8 while the total charged particle concentration increased by a factor of about 2. As in the NPF event (Fig. 2), this indicates that the production rate of cluster ions was not sufficiently fast to keep pace with the rapidly increasing PNC and that the particles were under-charged.

Pyrotechnic Display

The pyrotechnic display on 29th September 2012 began at 19:00 and ended at 19:25 and was continuous except for a short lull of about 5 min soon after 19:10. Fig. 5 shows the ion and particle concentrations during the time of the display. The wind remained westerly and showed a gradual increase from about 1 to 5 m/s during the course of the display. This was probably associated with thermal effects due to the extensive fireworks. The PNC was high in the hour before the display began (3×10^4 to 9×10^4 cm^{-3}) owing to various other activities that took place earlier as part of the festival. However soon after the inception of the fireworks, the PNC increased steeply from 5.5×10^4 to 1.35×10^5 cm^{-3} in 3 min. This is a rate of increase of PNC of 1.6×10^6 cm^{-3}/h, which is much larger than any other natural or anthropogenic process that we have observed at the site. During the lull in activity, the PNC dropped to 6 ×

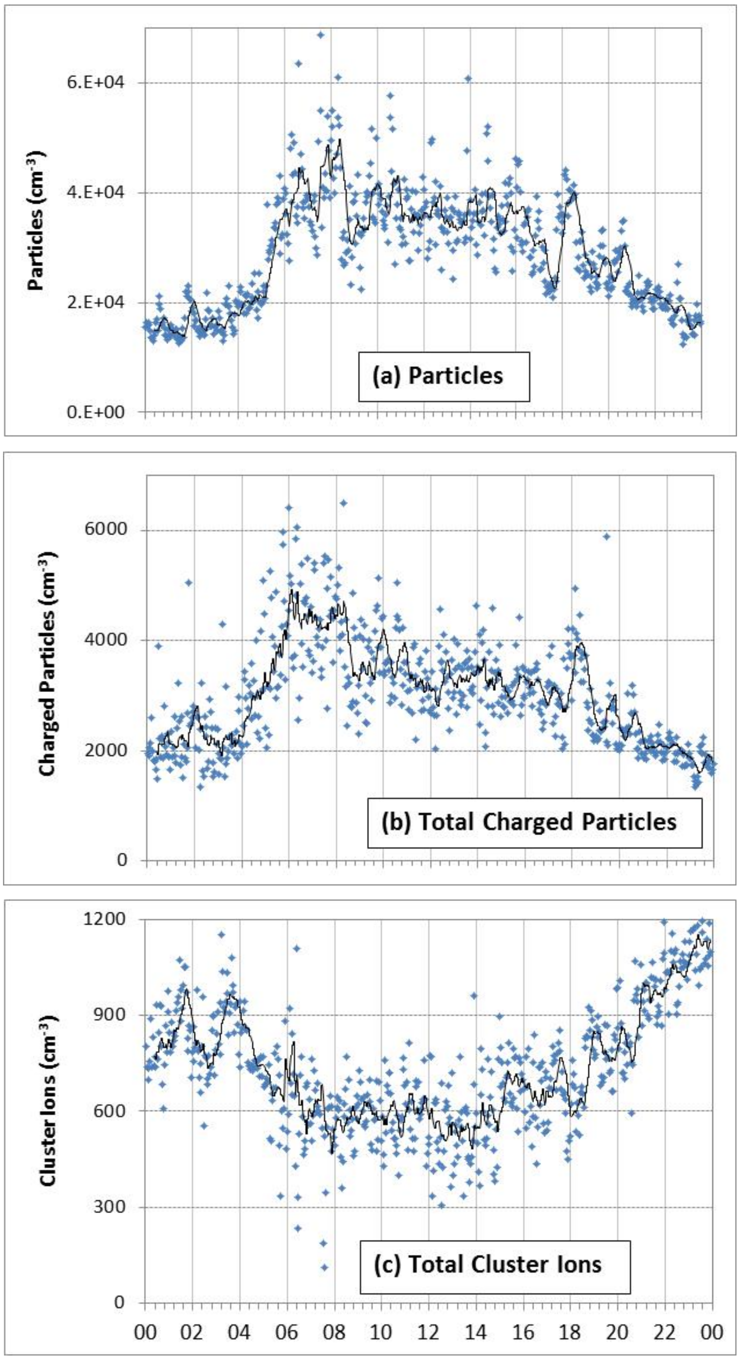

Fig. 1. Diurnal variation of particle and ion concentrations on 24 April - a typical day with no particle formation. The x-axis shows the time of day in hours. Each point is a 1 min average value and the curves represent 1-hour running averages.

10^4 cm^{-3} before rising again to 1.25×10^5 cm^{-3} in the next 6 min – a rate of increase of 6.5×10^5 cm^{-3}/h. The total CIC decreased steadily during the display from 400 cm^{-3} and reached zero near the end of the display. Fig. 6 shows the positive and negative CICs separately. In general, the ambient urban positive CIC is 10%–40% higher than the negative CIC at most times (Ling *et al.*, 2010). Soon after the pyrotechnics began, the negative CIC began to decrease rapidly reaching zero within 6 min. It then remained at zero for the next 18 min and climbed back up only once the display had ended. The positive CIC decreased more slowly but eventually reached zero for about 3 min near the end of the display. This difference is explicable in terms of the mobility of cluster ions in air. Negative ions have a higher mobility than positive ions in air and, therefore, a greater attachment coefficient (Horrak *et al.*, 2000; Retalis *et al.*, 2009). Thus, as the PNC increases, the negative CIC decreases faster than the positive CIC.

Unlike during NPF and bushfire events, the fractional increase in particle charge concentration was higher than the fractional increase in PNC. This suggests that, there is charge being generated during the pyrotechnic process and

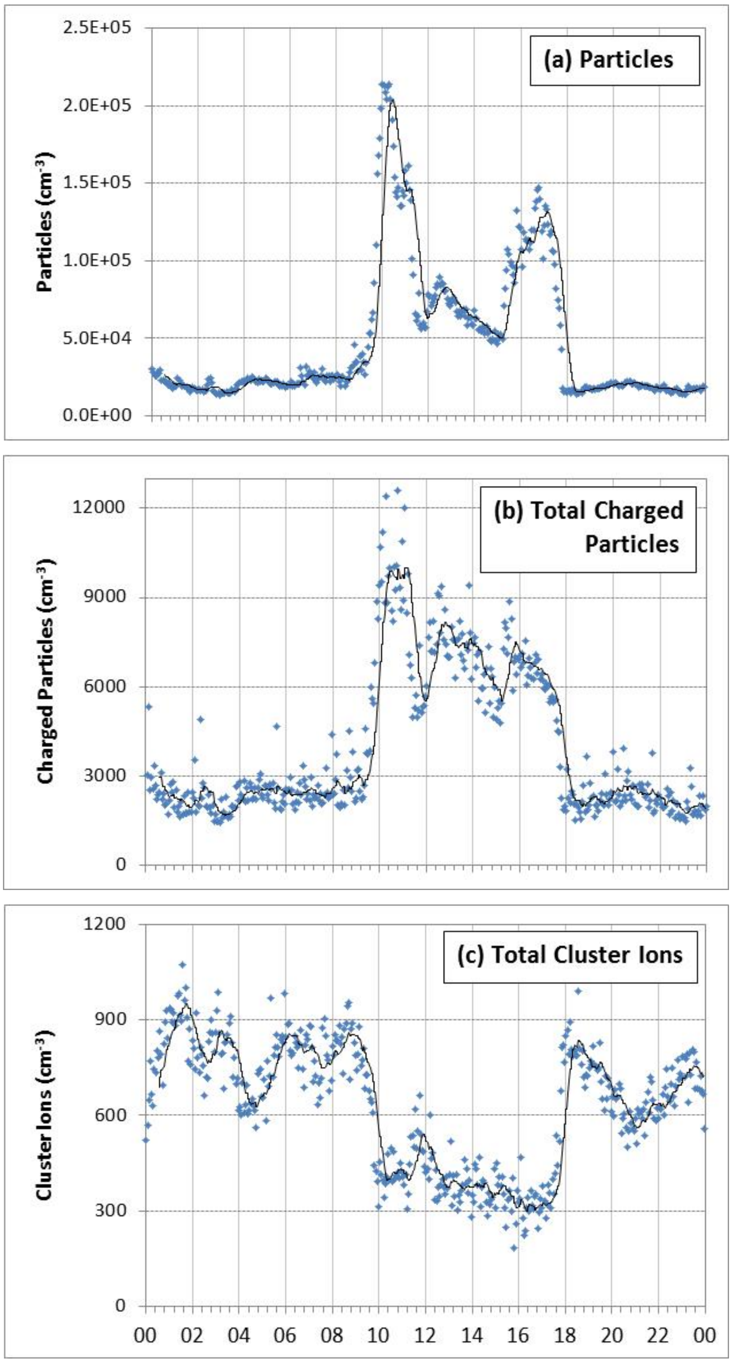

Fig. 2. Diurnal variation of particle and ion concentrations on 7 April, exhibiting a strong NPF event between 9:00 and 10:00. The x-axis shows the time of day in hours. Each point is a 1 min average value and the curves represent 1-hour running averages.

that at least some of the particles are charged at source. This is further confirmed by the sharp increase of both positive and negative CICs as soon as the fireworks ceased (Fig. 6). The total CIC increased from zero to over 600 cm^{-3} in 6 min, which is about twice the background value observed before the display (Fig. 5(c)). This shows that both cluster ions and charged particles are produced during pyrotechnic displays with positive and negative numbers being approximately the same. This is consistent with observations of other combustion processes and reflects what is observed in emissions from, for example, motor vehicles (Jayaratne *et al.*, 2010). In the present study, although the fireworks produced cluster ions, it is clear that they were rapidly and totally mopped up by the particles due to the very large number of particles that were produced in a very short time.

General Trends of CIC versus PNC

In Fig. 7, we chart the median values of the PNC against the total CIC of the various events described in this paper. Six types of events, excluding the 12 NPF events, are shown as

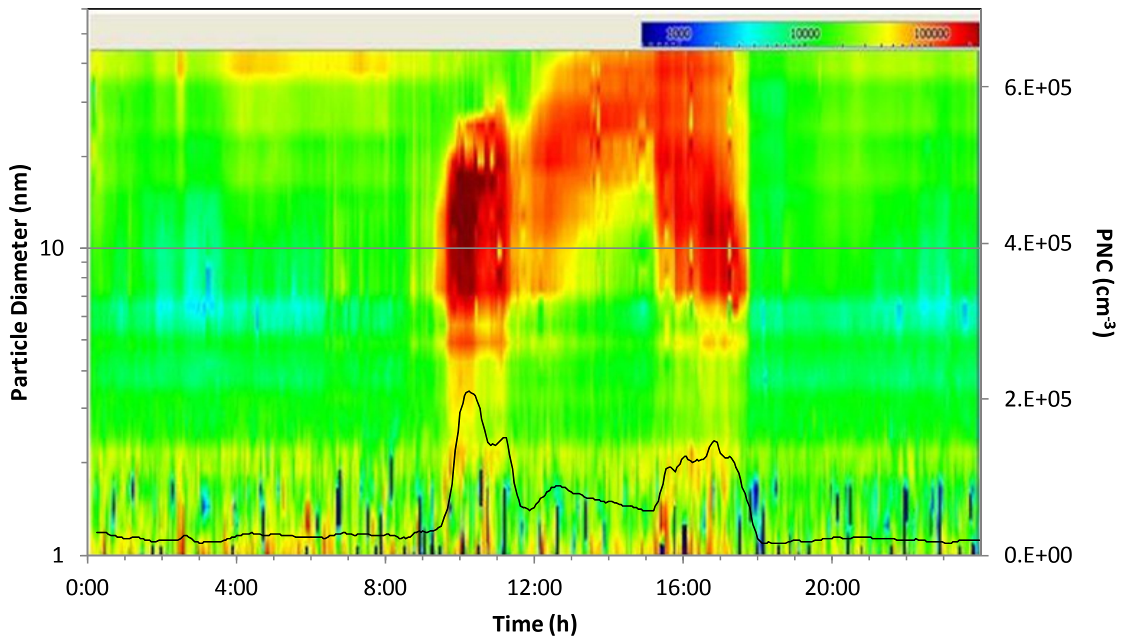

Fig. 3. NAIS spectragraph showing the temporal variation of the particle size distributions on 7 April. A strong NPF event occurs between 9:00 and 10:00. The curve at the bottom shows the PNC.

filled diamonds with the error bars representing the respective first and third quartile values about the medians. Urban Days and Urban Nights represent the mean concentrations between 6:00–18:00 and 18:00–6.00, respectively, on the days when there were no NPF events observed. We have included the clean rural site and the polluted roadside site. The latter point comprises measurements over a 30 min period near the curb of a busy motorway and the values are derived from Jayaratne *et al.* (2014). The pyrotechnic event between 19:00 and 19:25 on 29 September (Fig. 5) and the bushfire event on 18 December between 10:00 and 14:00 (Fig. 4) are included. Both these events gave relatively high PNCs and low CICs. The median value of the total CIC pertain to the entire period of the pyrotechnic display and, therefore, does not show the observed short term decrease to zero seen in Fig. 5(c). The total CICs at the clean rural site and during the bushfire episode showed the highest and lowest median values, respectively. Urban nights showed a higher total CIC than urban days because the mean PNC during the day was always greater than during the night, presenting a larger sink for the cluster ions. As expected, the widest variation in both total CIC and PNC were observed at the roadside owing to the highly variable nature of the emissions from passing vehicles (Jayaratne *et al.*, 2010). The median total CIC at the roadside was significantly higher than during the pyrotechnic display and the bushfire event. This is not surprising as we know that, together with particles, motor vehicles emit large concentrations of cluster ions (Jayaratne *et al.*, 2010). Despite these different processes, there is an overall approximately linear decrease of total CIC with increasing PNC, as evidenced by the broken line ($R^2 = 0.87$; $P < 0.05$). This is explicable in terms of the increasing probability of attachment of cluster ions to particles as the PNC increases.

On the same chart (Fig. 7), we have shown the 12 NPF events as open squares. The error bars are not shown in the interest of clarity, but did not exceed 100 cm^{-3} in any of the 12 events. The point at the far right is the short burst NPF event between 9:00 and 12:00 on 7 April where the PNC exceeded 2×10^5 cm^{-3} in less than 1 h (Fig. 2). Eight of the twelve points are clustered around total CIC = 600 cm^{-3} and PNC = 6×10^4 cm^{-3} and these represent the majority of NPF events. The other four NPF events showed higher PNC values resulting from strong growth and demonstrated clear ‘banana events’. As with the other six situations, the second broken line shows that the total CIC decreased with PNC but at a much lower rate. It is interesting to note that the median total CICs in the 12 NPF events were limited to a relatively narrow range between 300 and 700 cm^{-3}. However, all 12 CIC values were significantly greater than that observed during both the pyrotechnic and bushfire events, and greater than that at the roadside. There is a plausible explanation for these observations. Most of the particles in an NPF event are secondary volatile particles that are in the nanoparticle size range (Morawska *et al.*, 2009) while the particles produced by fireworks and bush burning consist of mostly primary soot that are much larger in size. Airborne measurements of PNC near biomass burning events in the Northern Territory, Australia (Ristovski *et al.*, 2010), showed that the median diameter of smoke particles were about 90 nm when the flight paths were close to the fires. At a few hundred km distance, the size increased to 130 nm due to coagulation. Considering that the fires in the present study were 100–200 km away, it is likely that the particles were well over 100 nm in size. Particle number size distributions measured in Leipzig,

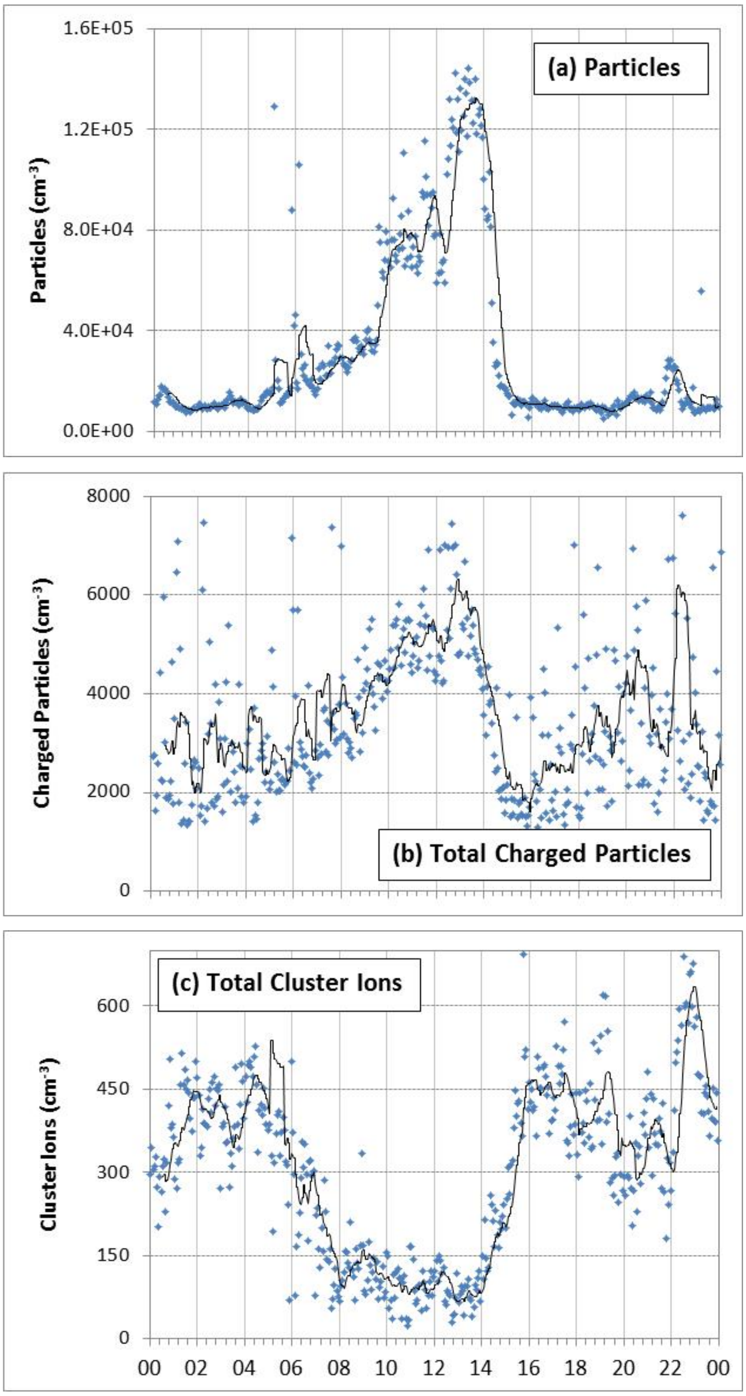

Fig. 4. Diurnal variation of particle and ion concentrations on 18 December - a typical bushfire event day. The x-axis shows the time of day in hours. Each point is a 1 min average value and the curves represent 1-hour running averages.

Germany, during the millennium fireworks 2000, showed a concentration maximum at a size of 100 nm shortly after midnight which was significantly greater than the value of 30 nm found at other times (Wehner *et al.*, 2001). Thus, at the same PNC, the surface area of the particles is much greater during the pyrotechnic display and the bushfire episode than during an NPF event. Moreover, larger particles are able to hold a higher charge than smaller particles (Wiedensohler, 1988). These effects would clearly enhance the attachment of clusters to particles, decreasing the CIC in the atmosphere. Motor vehicle emissions show a wider range of particle size including both secondary particles and primary soot (Morawska *et al.*, 2008) and, therefore, explains why the total CIC value of the roadside point lies between the NPF events and the pyrotechnic and bushfire points.

SUMMARY AND CONCLUSIONS

A study of seven different types of events in the atmosphere showed a general trend of decreasing total CIC

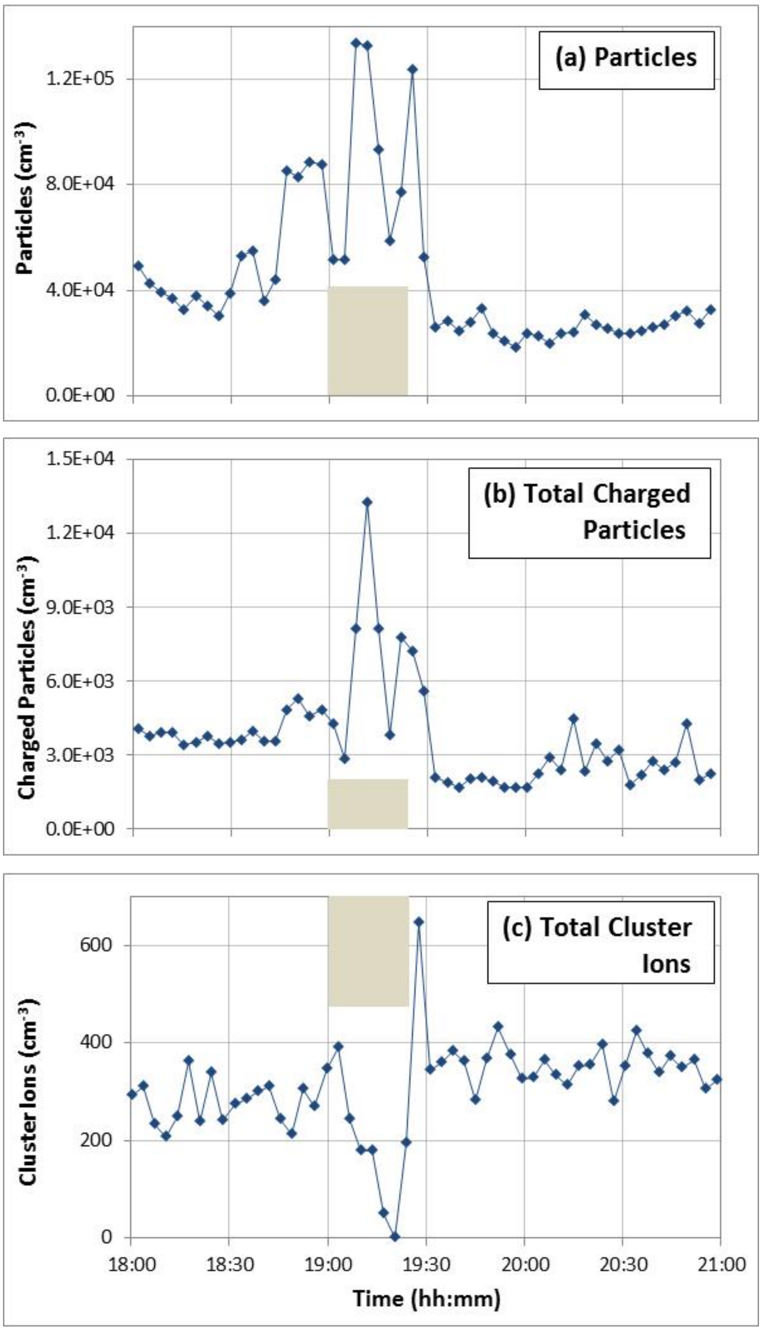

Fig. 5. Variation of particle and ion concentrations during the pyrotechnic display on 29 September. The display lasted from 19:00 to 19:25 and this time period is indicated by the shaded area in each graph.

with increasing PNC (Fig. 7). This result is in good agreement with previous studies that have shown that aerosol particles in the atmosphere can readily suppress cluster ions (Ling *et al.*, 2013). However, in certain cases, we observed widely different values of total CIC for the same PNC. For example, the PNC during the pyrotechnic display and the bushfire episode were both within the same range as that observed during the NPF events. However, the total CIC during each of these two events was significantly lower than that at every NPF event. In fact, during the pyrotechnic display, the total CIC was suppressed completely to zero for a few minutes. This is a novel observation. We attribute this to the rapid increase of PNC which allows the particles to mop up the cluster ions faster than they are produced. Also, during the bushfire episode, the total CIC fell to a value that was significantly lower than during the NPF events that showed the same PNC. This we attribute to the larger size of particles in the bushfire smoke than in the atmosphere during NPF events. Larger particles are able to mop up cluster ions more effectively than smaller particles.

Thus, our results suggest that, while CIC generally decrease with increasing PNC, rapid changes in PNC and larger particles also contribute significantly to the suppression of CICs in the atmosphere.

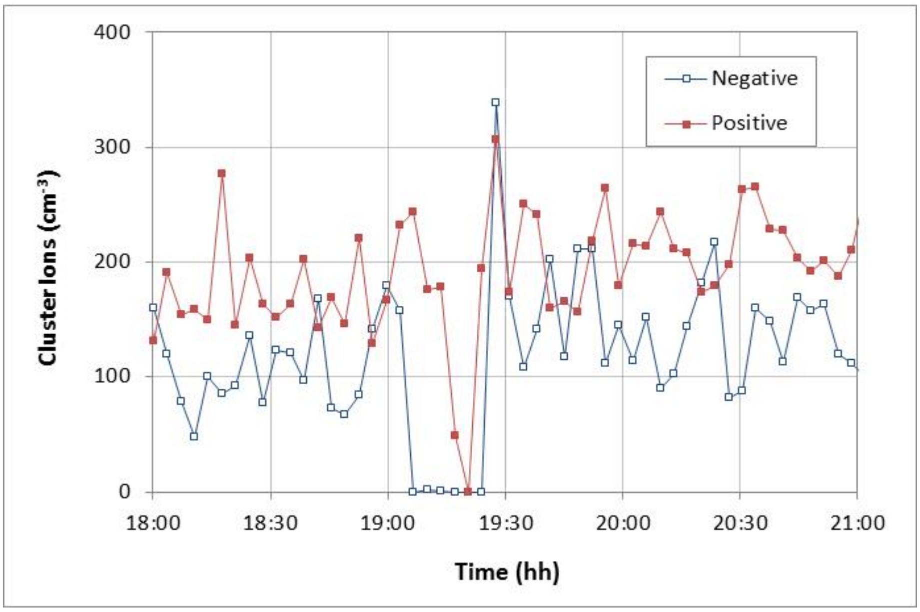

Fig. 6. Variation of negative and positive CICs during the pyrotechnic display on 29 September. The display lasted from 19:00 to 19:25.

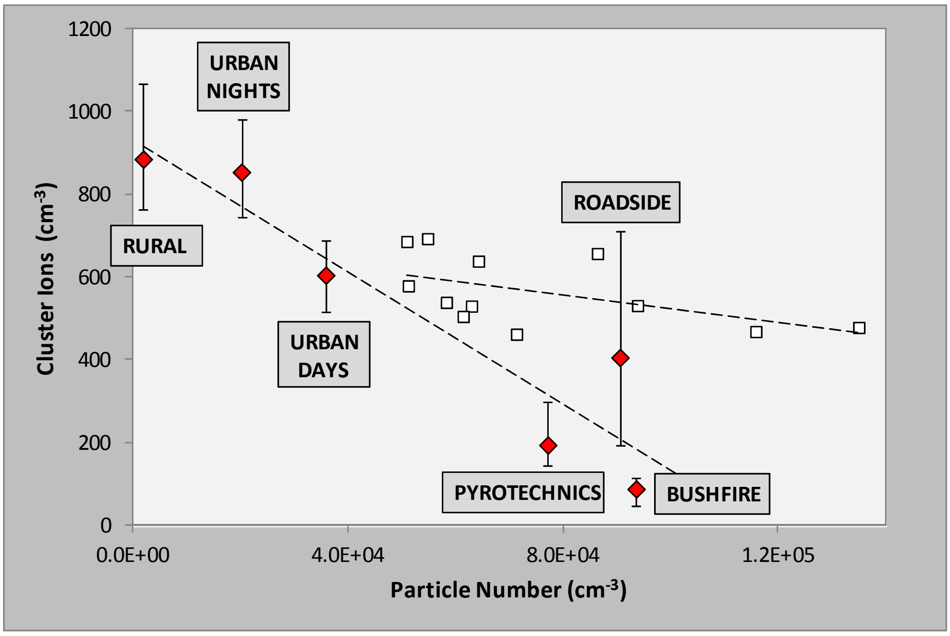

Fig. 7. Median values of the CIC plotted against the corresponding PNC for six situations (filled diamonds). The error bars represent the respective first and third quartiles. The 12 NPF events are shown as open squares. The broken lines present the best linear fits through the two sets of points.

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