Atmospheric Environment 190 (2018) 10–22

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

# Classification of the new particle formation events observed at a tropical site, Pune, India

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ARTICLE INFO

*Keywords:*
Classification of air ions
New particle formation
Mobility distribution
Secondary particles

ABSTRACT

A total number of 109 new particle formation events identified in the ion-mobility spectra measured with a Neutral Cluster and Air Ion Spectrometer in the mobility range of 3.16–0.00133 $cm^2 V^{-1} s^{-1}$ (Diameter* range 0.36–47.1 nm) at a tropical site at Pune, (18.53 °N, 73.85 °E, 573 m amsl) India from March 08, 2010–December 31, 2012 are classified based on their shape characteristics under four categories. Most of these events occurred in the morning hours of the pre-monsoon season during the hottest months (April and May) of the year. The meteorological conditions and the changes in ion characteristics associated with some typical events are examined. Average ion-mobility spectrum for the event days shows a minimum in the negative big cluster ion (diameter, 0.85–1.6 nm) concentration and two maxima in the positive intermediate (diameter, 1.6–7.4 nm) and large ion (diameter, 7.4–47.1 nm) concentrations as compared to the average spectrum for all days. Analysis of 7-days airmass back trajectories shows that since the only source of big cluster ions is through the growth of small cluster ions (diameter, 0.36–0.85 nm) the growth of small to big cluster ions is faster when the airmass approaches our site from the land. Further, the concentrations of positive intermediate and light large (diameter, 7.4–22 nm) ions is more when the airmass approaches from the Arabian Sea.

## 1. Introduction

[2]Atmospheric aerosols can be either injected into the atmosphere directly from land or sea surface - primary aerosols, or can be generated in the atmosphere by the gas-to-particle conversion processes - secondary aerosols. Atmospheric ions are actively involved in the formation of secondary aerosol particles (Hoppel, 1985: Yu and Turco, 2000; Kulmala et al., 2004; Gopalakrishnan et al., 2005; Yu, 2010; Tammet et al., 2014). Knowledge of the changes in size and mobility of air ions in the initial stages of their growth provides valuable information for understanding the formation and growth processes of aerosols. Knowledge of the process responsible for the growth of the newly formed particles to the size of cloud condensation nuclei where they can influence the Earth's radiation budget and cloud processes is much needed under different environmental conditions for estimating the atmospheric radiative forcing on the regional and global scales. Observations to investigate the formation and growth of ultrafine particles have been made under a variety of environmental conditions (Kulmala et al., 2004, 2013; Dunn et al., 2004; Laakso et al., 2004; Hirsikko et al., 2005; Siingh et al., 2005, 2011a; Tammet et al., 2006; Qian et al., 2007; Kamra et al., 2009, 2015a; Asmi et al., 2010; Manninen et al., 2010; Betha et al., 2013; Kanawade et al., 2014; Young et al., 2013; Bianchi et al., 2016; Huang et al., 2016). Measurement sites for such observations extend over large ranges of altitude, latitude and degree of air pollution. For example, such measurements have been made at different altitudes in the atmosphere (Lee et al., 2003; Bianchi et al., 2016), in the tropical, mid- and high-latitude and polar regions (Dhanorkar and Kamra, 1993a,b; Horrak et al., 1998; Siingh et al., 2007, 2011a,b; Kamra et al., 2009; Manninen et al., 2010; Jung et al., 2013), and in the remote marine, polluted marine, remote continental, rural and urban areas (Hoppel et al., 1994; Zhang et al., 2004; McMurry et al., 2005; Qian et al., 2007; Hirsikko et al., 2011; Gagné et al., 2011; Herrmann et al., 2014; Kanawade et al., 2014; Santos et al., 2015). Urban areas are an important source for the global aerosol CCN load and most of CCN come from the primary emission and only 10% from nucleation in the boundary layer (Merikanto et al., 2009). Nevertheless, the number of studies focusing on the behaviour of air ions and particularly their association with the NPF events in urban areas around the world is still somewhat limited (Manninen et al., 2010). The main emphasis in the past studies, some of which were based on the observations made over a

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[2] Diameter refers the “mass diameter” throughout the manuscript.

https://doi.org/10.1016/j.atmosenv.2018.07.025
Received 5 January 2018; Received in revised form 7 July 2018; Accepted 10 July 2018

period of several years, was to study the nucleation events for the formation of new particles.

A variety of different mechanisms such as the binary (Kulmala and Laaksonen, 1990; Viitanen et al., 2008), ternary (Kulmala et al., 2000; Viitanen et al., 2008), ion-induced (Yu, 2001) and ion-mediated nucleation (Yu and Turco, 2000; Svensmark et al., 2007; Kirkby et al., 2011; Nagato and Nakauchi, 2014; Riccobono et al., 2014), and the nucleation mechanisms involving organic vapors (O'Dowd et al., 2002a) or iodine (O'Dowd et al., 2002b) have been proposed to be responsible for the nucleation of the newly formed particles. Moreover, vapours that participate in different steps of nucleation are most likely different from those that participate in the growth of particles (DallÓsto et al., 2018). A satisfactory dominant mechanism for the nucleation and growth of the particles at most of the locations has remained elusive. Moreover, such measurements and knowledge of the mechanisms responsible for the formation and growth of the particles in tropics are scarce and are much needed for understanding the aerosol effects on the climate and climatic change.

Formation of new particles in nucleation events mostly occurs in “bursts” (Horrak et al., 1998) which have been classified in different categories based on their shapes. For example, Hirsikko et al. (2007) and Vana et al. (2008) have classified the New Particle Formation (NPF) events in different categories based on their observations made at mid-latitude stations at Hyytiala (boreal forest) and Mace Head (west coast of Ireland), respectively. To our knowledge, such a classification has never been reported for the NPF events observed at a tropical site. In the present work, we classify the NPF events observed over a period of ~3 years under four categories and report on the seasonal distribution of each category of the events. The airmass back trajectories and the diurnal variations and changes in the ion size and mobility characteristics associated with each category are also examined and reported. We also compare the characteristics of the average mobility spectra (i) on the event days and on all days, and (ii) when the airmass is reaching our measurement site from the ocean or land are also compared. Towards this objective, we have measured the ion mobility spectra during March 2010 to December 2012 at Pune (18° 31′ N, 73° 55′ E, 560 m above mean sea level) with a Neutral Air Ion Spectrometer (NAIS) in the mobility range of 3.16–0.00133 $cm^2 V^{-1} s^{-1}$ which corresponds to the particle size range of 0.36–47.1 nm diameter.

## 2. Instrumentation and observation site

Measurements of both positive and negative ions in the mobility range of 3.16–0.00133 $cm^2 V^{-1} s^{-1}$ (diameter range 0.36–47.1 nm) have been made with a Neutral Air Ion Spectrometer (NAIS) of Airel Ltd, Estonia (Mirme et al., 2007). The NAIS records mobility spectra for positive and negative ions in a 5-min period, based on 200-s of sample air and 100-s offset-level measurements. The entire mobility distribution of the atmospheric ions can be divided in two main classes, (i) cluster ions (mobility $> 0.5\,cm^2 V^{-1} s^{-1}$; diameter $< 1.6$ nm) and (ii) charged nanoparticles; mobility $< 0.5\,cm^2 V^{-1} s^{-1}$; diameter $>$ 1.6 nm) (Horrak et al., 2003). These two classes of ions have been further divided into five independent categories based on their size and mobility, viz, small cluster ions (mobility 3.16–1.3 $cm^2 V^{-1} s^{-1}$, size 0.36–0.85 nm diameter), big cluster ions (mobility 1.3–0.5 $cm^2 V^{-1} s^{-1}$, size 0.85–1.6 nm diameter), intermediate ions (mobility 0.5–0.034 $cm^2 V^{-1} s^{-1}$, size 1.6–7.4 nm diameter), light large ions (mobility 0.034–0.0042 $cm^2 V^{-1} s^{-1}$, size 7.4–22 nm diameter), heavy large ions (mobility 0.0042–0.00133 $cm^2 V^{-1} s^{-1}$, 22–47.1 nm diameter) (Horrak et al., 2000).

The NAIS was placed inside a laboratory on the second floor of a 3 - storey building of Indian Institute of Tropical Meteorology, Pune. The inlet for the NAIS consisted of a metallic pipe which has a length of 40 cm and a diameter of 3 cm. To minimize the particle loss, the pipe had no sharp turns and was fixed in a cutout ($2 \times 7\,m^2$) of the building, with its open end facing down at a height of ~6 m above ground level. Since the length of the pipe for inlet was less than that recommended (50 cm) by the manufactures, any loss of the small ions to the pipe wall was taken as negligible. The inlet pipe was shielded from the atmospheric electric field by vertical walls of the building on three sides with one side open and a projected roof on the top, but still was well-ventilated in the atmosphere.

The institute is located on the outskirts of a moderately big town with a population of ~9 millions. The institute campus is located in a valley surrounded by 500–800 m high hills on three sides. A 12 - meter wide road with moderate traffic runs in the east-west direction about 100 m away from the institute building. A small canteen and a residential area are about 200 m and 400 m away, respectively, from the site of measurement. The area is covered with cotton soil. The unique geographical location of Pune in south-west India makes it a suitable site for detailed studies of the rich mixtures of ions and aerosols transported from deserts, Indo-Gagantic Plaine and the Arabian sea in addition to the local emissions. A detailed description and the photograph of inlet pipe and the surrounding of the measuring site are given in Kamra et al. (2015a).

## 3. Criteria for the classification of the new particle formation events

Based on the visual inspection of the shape characteristics of the new particle formation (NPF) events (henceforth called events) during our observation period at Pune, we have classified them in different categories viz. Type Ia (Truncated banana), Type Ib, Type II (Patch type), Type III (Inverted cup type), type IV (Undefined type), Type V (Non-event). To complete the discussion and avoid the need of readers to often refer back to our earlier works, a short description and an example of each of the periods of enhanced concentration of ions and aerosol (PECIA) and rain-induced events are also added. We also report typical examples of each event and the meteorological conditions and ion characteristics changes occurring on such event days. An analysis of the hourly changes in dn/dlogD for every single event in our observations revealed that most of the events examined showed more or less similar qualitative trends in the size distribution curves during events of the same category. Trends in the features described in the examples of different categories of events are followed in a majority of cases in a particular category. However, significant deviations from these trends do exist in some cases. It is difficult, however, to quantify the deviations as the type of deviations differ over different ion size ranges. So these trends in features cannot be generalized but need be taken only as a guideline.

### 3.1. *Type Ia (Truncated banana type)*

This type of event occurred mostly in the morning hours between 0830 and 1300 LT at our site. In these NPF events, a distinct new mode of 4–6 nm diameter particles appeared in the size range of intermediate ions. The appearance of these particles and their growth to larger sizes continued for 3–4 h, which indicates the homogeneity of the airmass. The formation and growth of new particles visualized in the number size distribution plots in these events resembled a banana shape (Fig. 1a). However, these were characteristically different from the traditional banana type events observed at other locations (Vana et al., 2008; Laakso et al., 2008; Lehtipalo et al., 2010; Manninen et al., 2010), in the respect that they neither showed any simultaneous burst of small ions nor any persistent growth of ions of < 4–6 nm diameter which are typical features of banana type NPF events. However, these NPF events followed a banana shape for the ion growth from 4 - 6 nm–47.1 nm diameter. Formation of these particles may first start as molecular clusters at the top of the surrounding hills where solar radiation first hits the ground in the morning (Kamra et al., 2015a; Gautam et al., 2017). Subsequently, these newly - formed particles along with other aerosol particles, radioactive emanation and trace