



# Characteristics of Aerosol Size Distributions and New Particle Formation Events at Delhi: An Urban Location in the Indo-Gangetic Plains

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Accurate information about aerosol particle size distribution and its variation under different meteorological conditions are essential for reducing uncertainties related to aerosol-cloud-climate interaction processes. New particle formation (NPF) and the coagulation significantly affect the aerosol size distribution. Here we study the monthly and seasonal variability of aerosol particle size distribution at Delhi from December 2011 to January 2013. Analysis of aerosol particle size distribution using WRAS-GRIMM reveals that aerosol particle number concentration is highest during the post monsoon season owing to the effect of transported crop residue and biomass burning aerosols. Diurnal variations in number concentration show a bimodal pattern with two Aitken mode peaks in all the seasons. Monthly volume size distribution also shows bi-modal distribution with distinct coarse and fine modes. NPF events are observed less frequently in Delhi. Out of 222 days of WRAS data, only 17 NPF events have been observed, with higher NPF frequency during summer season. Growth rate of the nucleation mode of NPF events vary in the range 1.88–21.66 nm/h with a mean value of  $\sim 8.45 \pm 5.73$  nm/h. It is found that during NPF events the Aitken and nucleation mode particles contribute more to the number concentration. Simultaneous measurement of UV flux and particulate matter (PM<sub>10</sub> and PM<sub>2.5</sub>) have also been done along with particle number size distribution measurement to understand the possible mechanisms for NPF events over the study location.

**Keywords:** aerosol size distribution, nucleation, Aitken mode, new particle formation, particle growth rate

## INTRODUCTION

The size distribution of atmospheric aerosol particles are known to have plausible effects on radiation budget through direct interaction (Warner and Twomey, 1967; Liou, 1992; Andreae et al., 2004; Dusek et al., 2006); cloud characteristics by affecting cloud condensation nuclei, CCN (Ramanathan et al., 2001; Andreae et al., 2004) and droplet number density (Rosenfeld et al., 2008; Zhang et al., 2011); visibility (Hand et al., 2002; Bäumer et al., 2008; Singh and Beegum, 2013); communication (Ricklin et al., 2006); human health (Harrison, and Yin, 2000; Valavanidis et al., 2008) and

agriculture (Chameides et al., 1999). Although, the climatic and human health implications of aerosol have drawn great attentions in the last 2 decades (Lighty et al., 2000; Bergin et al., 2001; Singh and Dey, 2012; Boucher et al., 2013; Gogoi et al., 2021), there are still scarcity of high-resolution (both spatial and temporal) data on aerosol particle size distribution at distinct geographic parts of the world. Geographical and seasonal differences significantly influence the aerosol particle size distribution (Kompalli et al., 2014). Especially, over the highly heterogeneous geographic regions of South Asia, accurate and improved understanding of aerosol size distribution is highly essential to understand distinct sources, transformation and growth processes of aerosols.

While primary sources are dominant contributors to the abundance of atmospheric aerosols, secondary aerosol production is unique in altering aerosol size distribution pattern, thus changing their dynamical properties. Depending on the abundance of precursor gases, solar radiation and pre-existing particle concentrations (Kulmala et al., 2001; Ueda et al., 2016), gas to particle conversion (i.e., new particle formation - NPF) occurs leading to the formation of secondary aerosols (Kulmala et al., 2004; Dal Maso et al., 2005; Seinfeld and Pandis, 2016). Meteorology and the boundary layer dynamics also impact the NPF events frequency and formation mechanism (Kuang et al., 2010; Wehner et al., 2010). NPF events are mostly reported during day time, mainly related to the photochemistry formation theory (Kuang et al., 2008; Metzger et al., 2010). Sulphuric acid is considered as one of the main contributor to the NPF events, which in turn is produced by the photooxidation of sulphur dioxide and OH radicals (Lee et al., 2019). Recent study by Jokinen et al. (2017) showed the significance of photochemistry in NPF, by depicting the decrease in UV radiation followed by the decrease in concentration of  $\text{H}_2\text{SO}_4$  and nitrogen containing highly oxidized compounds during a partial solar eclipse, resulting in the inhibition of NPF events over the study site.

Delhi has a population of more than 1 million (Statistical Abstract of Delhi-2014) and has a large number of daily traffic on the roads. Two coal power plants (Rajghat and Badarpur) in the vicinity and the heavy industrial activities and high traffic intrusion account for the increased pollution events in Delhi (Chowdhury et al., 2017). The city, which comes under the Indo-Gangetic Plains (IGP), has a unique geographical and meteorological conditions. The IGP alluvial plains, the Thar Desert and the Aravalli Hills surround Delhi in north and west respectively and accounts for this uniqueness. Delhi encounters dust storm events during April-June and biomass burning events during the October-November months (Singh et al., 2005; Singh et al., 2010; Mishra and Shibata, 2012; Lodhi et al., 2013; Mishra et al., 2014).

Owing to the highly polluted scenario, a number of studies have been done on aerosol size distribution and NPF in Delhi (Mönkkönen et al., 2005; Sarangi et al., 2015; Pant et al., 2016). Increased concentration of  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$  particles are reported over Delhi during winter and post monsoon season (Mukherjee et al., 2018; Bhandari et al., 2020). Apart from particulate matters, gas species such as nitrates and sulphates are also reported in high concentration over the study location (Bhandari et al., 2020)

making conducive environment for chemical transformation and new particle formation events over Delhi. However, a systematic seasonal analysis of aerosol particle size distribution and NPF events are scarce over the region. The main aim of this paper is to recognize the monthly and seasonal variation in the number concentration and size distribution of the aerosol particles and see how the particle modes vary over the span of a year. Also, we present here the analysis on observed nucleation events during the time period. Important parameters related to NPF events such as growth rate and condensation sink have also been estimated during different seasons. This study also tries to enumerate some of the factors contributing to the observed NPF events during summer and winter season.

## DATA AND METHODOLOGY

### Measurement Site and Instrumentation

The aerosol particle size distribution measurements are carried out using Wide Range Aerosol Spectrometer (WRAS) located at CSIR-National Physical Laboratory (CSIR-NPL), Delhi (28.64° N, 77.17° E) during December 2011 to January 2013. The system comprises of a Scanning Mobility Particle (SMPS + C) and an Optical Particle Counter (OPC). The GRIMM SMPS + C system consists of a high resolution condensation particle counter (CPC) attached to the GRIMM “Vienna Type” M-DMA (Monodisperse Differential Mobility Analyser). DMA classifies the particles according to its electrical mobility which is then measured using a CPC. Both components together measure in the ultrafine size range from 5 to 350 nm. OPC which work on the light scattering technology for particle counting gives the particle size distribution in 31 channels ranging from 250 nm to 32  $\mu\text{m}$  (Grimm and Eatough, 2009). Semiconductor laser serves as the light source for OPC. Thus integrating both, SMPS + C and OPC data using a GRIMM software, the WRAS system gives the particle size distribution in 72 channel ranging from 5.5 nm to 32  $\mu\text{m}$ . There is an overlapping region from 250 to 350 nm due to the integration of SMPS and OPC, here we take the average number concentration for the analysis. A Nafion dryer is used inside the sampling probe in order to avoid the loss of volatile components and dehumidify the sample during the measurement period. The GRIMM-WRAS data is supposed to have an uncertainty in total flow rate up to 5% as per manufacturer specification. SMPS is reported to have an uncertainty of less than 10% for particles in the size range of 20–200 nm, while for particles above and below this range uncertainty increases (Wiedensohler et al., 2012). Uncertainty of OPC is estimated to be about 9.9% (Grimm and Eatough, 2009). The detailed description and principle of the instrument are discussed elsewhere (Grimm and Eatough, 2009). The system also has an integrated meteorological sensor which provides temperature, relative humidity and wind vector data.

The incoming global (direct + diffused) solar radiation were measured on a horizontal surface in the UV (280–400 nm) range using the Kipp & Zonen CUV-4 radiometer respectively. The radiation flux measurements have an estimated experimental error of 3% in the shortwave and <10% in the UV range.

More details about the radiation flux measurements are reported elsewhere (Bano et al., 2013). Ambient SO<sub>2</sub> data is obtained using the self-calibrated Horiba APSA-370 SO<sub>2</sub> analyzer which uses Ultraviolet Fluorescence (UVF) method as the operating principle for measurements. The detailed working principle is reported elsewhere (Suneja et al., 2019).

Continuous sampling was done from December 2011 to January 2013, however, some data gaps exist on a few occasions whenever technical errors were encountered during the observation. A total of 222 days' data has been studied and analyzed seasonally, categorized as, winter (DJF), summer (MAMJ), monsoon (JAS) and post monsoon (ON). Considering the regional climatic conditions June has been grouped into summer as south west monsoon hits Delhi by the end of June or early July. The instrument provides aerosol particle size distribution at a span of 5 min interval. Percentage data availability for winter, summer, monsoon and post-monsoon are ~36%, ~49%, ~35 and ~63% of the total days respectively. The solar flux data was obtained in Wm<sup>-2</sup> every 2 min, during the observation period and averaged hourly and seasonally.

## Identifying NPF Events

During the preliminary stages of the analyses the data has been flagged and removed if any discrepancy in the instrument or measurement is noted. Out of the entire experimental period, a total of 222 days of data has been found proper and the same has been used for further analysis. NPF events have been identified from the collected data sets based on the following criteria, as also suggested by (Dal Maso et al., 2005): 1) A distinctly new mode of particles must appear in the size distribution; 2) the mode must start in the nucleation mode; 3) the mode must prevail over a time span of few hours; 4) the new mode must show signs of growth. Further, the NPF events have been identified according to the following scrutiny techniques. 1) The visual identification of the contour plots of particle size distribution over the 24 h period. 2) The variation of mode diameter across 24 h period. 3) Individual mode analysis of the size distribution at each 5 min interval.

In this study, we have classified the modes into three categories, Nucleation mode (5–30 nm), Aitken mode (30–100 nm) and accumulation mode (100–1,000 nm) (Ueda et al., 2016). The data has been classified into event days, non-event days, and unidentified event days according to a number of criteria discussed in various peer reviewed papers (Dal Maso et al., 2005; Kanawade et al., 2014; Kamra et al., 2015; Tröstl et al., 2016). Based on this, three categories of events (classifications) emerge with their distinct features. First, 1) *NPF Event days*, which are those days in which there is a sudden outburst of number concentration in the size range between 5 and 30 nm for a prolonged time period of 2–3 h (Kanawade et al., 2014). Another category, 2) *Non Event days* are those for which no nucleation and particle growth is observed in these days. The contour plots show no particular trend or variations in the number concentration. Finally, third category 3) *Unidentified events days* are the ones where we are unable to deduce the features of the number concentration from the contour plot and other scrutiny methods due to high level of noises in the data or

the gaps in the data. This can be due to the background pollution and the haze conditions prevailing in the atmosphere.

## Growth Rate Calculations

The rate of change of diameter of the particle ( $dD_p/dt$ ) is termed as growth rate (GR), where  $D_p$  is the diameter of the particle and  $t$  is time (Tröstl et al., 2016). The growth rate can be found out from the linear regression analysis of the mode diameter ( $D_p$ ) with time (Tröstl et al., 2016; Salimi et al., 2017). The slope of the regression gives the growth rate. The linear regression equation is given by

$$Dp = a_0 + a_1t, \quad (1)$$

where  $a_0$  is the intersect at the  $y$  axis and  $a_1$  is the slope of the line pointing to the growth rate.

## Condensation Sink

Condensation sink (CS) is the scavenging speed of the gaseous molecule due to the condensation onto particles. It mainly depends on the particle size and the condensation (Ueda et al., 2016), and the equation is given by Kulmala et al. (2001) as

$$CS = 2\pi D_v \sum_i D_{p,i} \beta_{m,i} N_i, \quad (2)$$

$D_v$  is the diffusion coefficient of H<sub>2</sub>SO<sub>4</sub> and is assigned as 0.104 cm<sup>2</sup> s<sup>-1</sup> (Kanawade et al., 2014).  $D_{p,i}$  and  $N_i$  is the center particle diameter and number concentration of the size class  $i$  respectively.  $\beta_{m,i}$  denotes the size dependent transition correction factor given by Fuchs and Sutugin (1971) and is expressed as

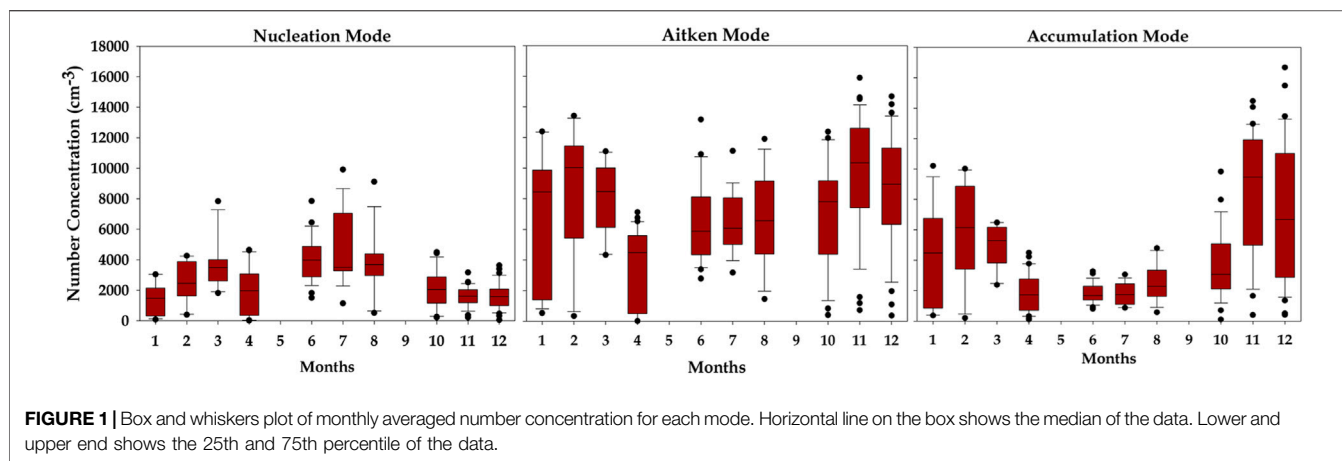
$$\beta_{m,i} = \frac{1 + K_{n,i}}{1 + 0.377K_{n,i} + \frac{4}{3}\alpha^{-1}K_{n,i} + \frac{4}{3}\alpha^{-1}K_{n,i}^2}, \quad (3)$$

where  $K_{n,i}$  is the Knudsen number for the size class  $i$  and the mass accommodation coefficient ( $\alpha^{-1}$ ) is assumed as unity.

## RESULTS AND DISCUSSION

### Aerosol Particle Concentrations in Different Size Regimes

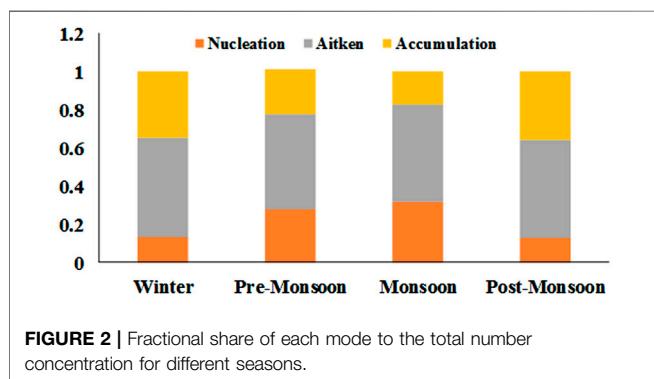
Figure 1 shows the box and whiskers plot of monthly averaged aerosol particle number concentration for different modes. The maximum aerosol number concentration for an individual day is recorded in December with a value of  $16.67 \times 10^3$  cm<sup>-3</sup>. The mode wise analysis of number concentration shows that Aitken mode (30–100 nm) dominates in the study region throughout the year with a maximum mean concentration during the post monsoon season ( $9.6 \times 10^3$  cm<sup>-3</sup>). Accumulation and nucleation mode particles are minimal compared to the Aitken mode particles during the entire study period at the sampling site. However, seasonal patterns showed variations in both the modes. Nucleation mode particles are high in number than accumulation mode particles during summer and monsoon seasons, whereas accumulation mode particles dominate during the other two seasons in comparison with nucleation mode. High mean concentration of nucleation mode particles ( $4.67 \times 10^3$  cm<sup>-3</sup>)



**FIGURE 1** | Box and whiskers plot of monthly averaged number concentration for each mode. Horizontal line on the box shows the median of the data. Lower and upper end shows the 25th and 75th percentile of the data.

**TABLE 1** | Seasonal mean number concentration for different particle modes at Delhi.

Seasons	$N_{5-30}$ ( $10^3 \text{ cm}^{-3}$ )	$N_{30-100}$ ( $10^3 \text{ cm}^{-3}$ )	$N_{100-1000}$ ( $10^3 \text{ cm}^{-3}$ )	$N_{5-1000}$ ( $10^3 \text{ cm}^{-3}$ )
Winter	$1.85 \pm 0.65$	$7.76 \pm 1.51$	$5.35 \pm 1.58$	$15.0 \pm 3.53$
Pre Monsoon	$2.73 \pm 1.54$	$5.17 \pm 2.76$	$2.59 \pm 1.57$	$10.5 \pm 5.50$
Monsoon	$4.06 \pm 0.53$	$6.73 \pm 0.26$	$2.26 \pm 0.41$	$13.0 \pm 0.16$
Post Monsoon	$1.83 \pm 0.38$	$8.09 \pm 1.48$	$5.90 \pm 3.17$	$15.8 \pm 4.26$



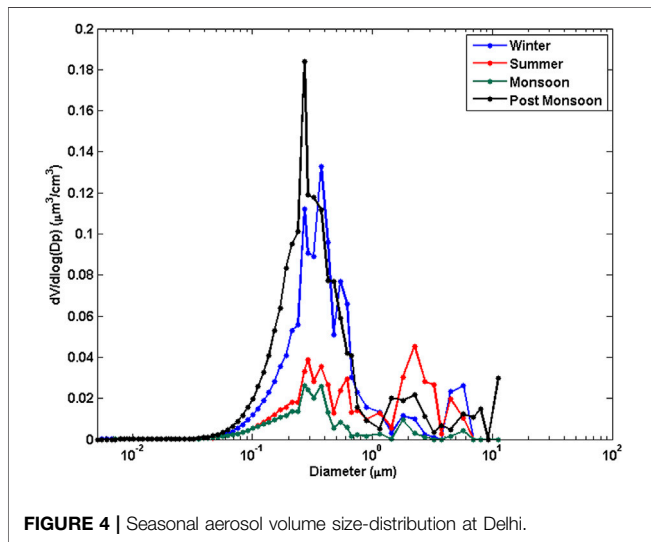
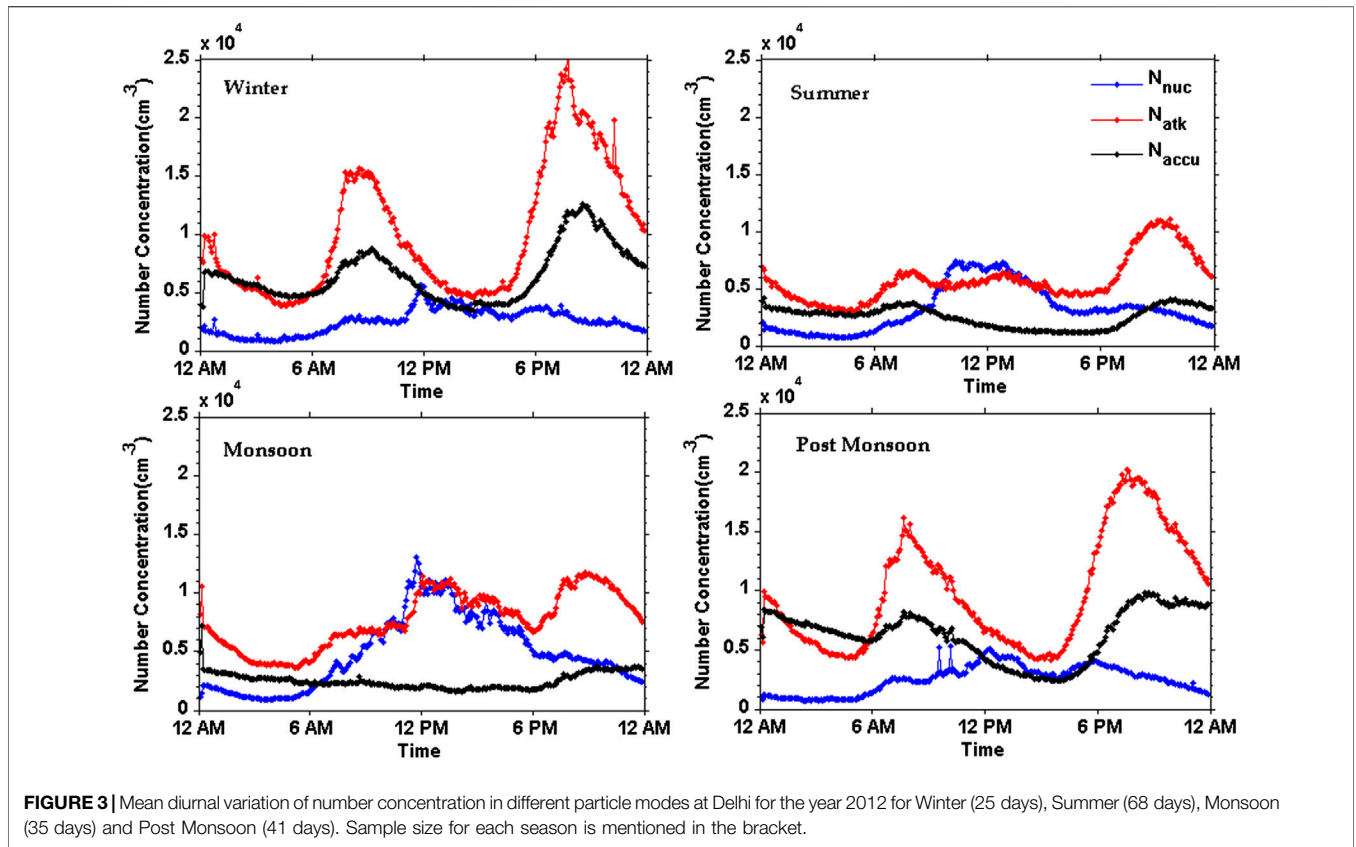
**FIGURE 2** | Fractional share of each mode to the total number concentration for different seasons.

are noticed during July, when the minimum mean concentration of accumulation mode particles ( $1.79 \times 10^3 \text{ cm}^{-3}$ ) are registered. Wet deposition of accumulation mode particles could be a plausible reason for the decreased concentration of accumulation mode particles and increased concentration of nucleation mode particles in the monsoon season. Heat map analysis of aerosol particle size distribution revealed a considerable shift in the size of particle as it moves from winter to pre monsoon.

The seasonal mean number concentrations of aerosol particles in different modes are tabulated in **Table 1**. Here,  $N_{5-30}$  indicates number concentration in nucleation mode,  $N_{30-100}$  in Aitken mode and  $N_{100-1000}$  in accumulation mode. Highest average total number concentration of  $15.80 \pm 4.26 \times 10^3 \text{ cm}^{-3}$  is noted during the post monsoon season, this can be attributed to the biomass burning events during the post monsoon seasons from nearby agrarian states like Punjab and Haryana. Mean total number

concentration for the entire experimental period is found to be  $14.08 \pm 7.60 \times 10^3 \text{ cm}^{-3}$ . Fractional share of each mode to the total number concentration is shown in **Figure 2**. The highest concentration of nucleation mode particles were observed during monsoon season with the mean number concentration reaching up to  $4.06 \pm 0.53 \times 10^3 \text{ cm}^{-3}$  with a fractional share of 0.315. The maximum number concentration of accumulation mode particle was found to be  $5.90 \pm 3.17 \times 10^3 \text{ cm}^{-3}$  in post monsoon season with a fractional share of 0.36. A considerable increase in the accumulation mode particles is seen during post monsoon and winter season as the major contributor for accumulation mode particles come from biomass burning activity (Chen et al., 2017; Ghosh et al., 2019).

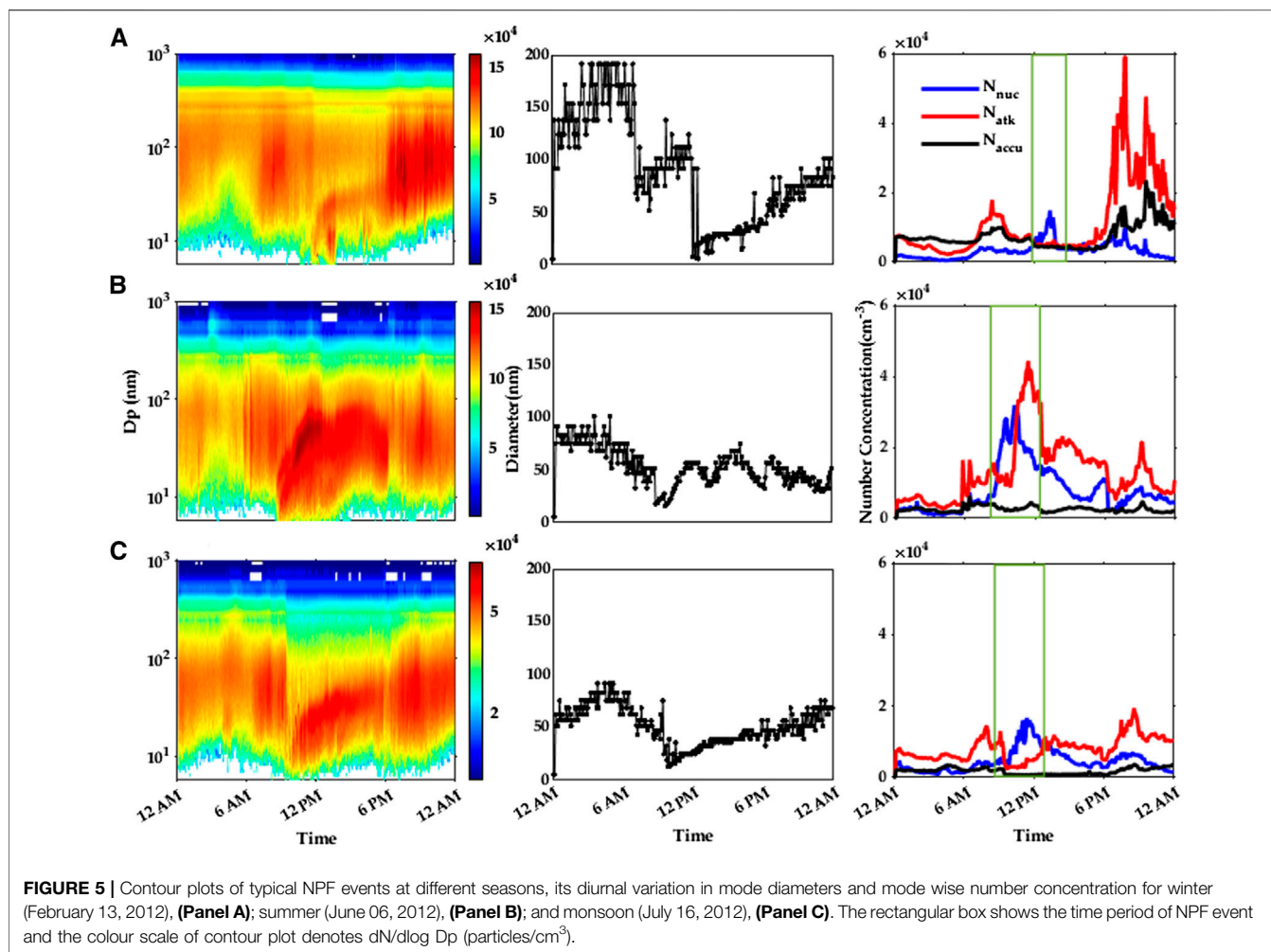
**Figure 3** depicts the diurnal variation in mean number concentration for different seasons. Aitken mode is the primary peak and follows a bimodal pattern for all the seasons. Starting from a gradual increase from early morning Aitken mode reaches a peak value at about 09:00–10:00 AM and then decreases. Aitken mode again shows an increase in concentration during the evening hours which later dies out at late night. This high particle concentration during the morning and evening hours is associated with the high rush of traffic during these hours. Among the two Aitken mode peaks observed, evening peak is seen to be prominent with higher concentration of Aitken mode particles, which may be due to the boundary layer dynamics associated with the diurnal meteorology. The decrease in the concentration of Aitken mode particle after the morning peak hours can be attributed to the mixing of air due to increased convection as the temperature increases and the increased concentration at the evening hours may be attributed to the stable layer formation during the evening, trapping the pollutants



inside the boundary layer (Mönkkönen et al., 2005). The pattern shows a similar trend in all seasons except for monsoon season, where the peak concentration is observed in the midday. The morning peak Aitken mode is succeeded by a small increase in nucleation mode particles during midday on few days and some of it later grows to nucleation events which are discussed in the following session.

Figure 4 shows the aerosol volume size distribution for different seasons. Bimodal pattern is observed for volume size distribution, characterizing fine (<1 μm) and coarse mode (>1 μm). For volume size distribution fine mode dominates and peaks at 0.38 and 0.27 μm for winter and post monsoon respectively. Coarse mode dominates during summer season and peaks at 2.25 μm. Increase in coarse mode particles during summer could be due to the high dust episodes (Mishra and Shibata, 2012) in the study location. Fine mode dominates for monsoon with a tiny peak at 0.38 μm. High volume concentration is noticed during the winter and post monsoon seasons with maximum volume concentration during November. Increased concentration of accumulation mode particles due to the surge in biomass burning activities could be the possible reason for the high volume concentration during post-monsoon season. It has also been noted that coarse mode particles are less predominant in these seasons and constitute a very feeble peak in the size distribution.

There haven't been many studies on long term measurements of aerosol number size distribution over Delhi. Hyvärinen et al. (2010) studied Gual Pahari, a semi urban area near Delhi and reported increase in nucleation and Aitken mode particles in summer and monsoon seasons, whereas accumulation mode was seen dominant during winter season. They also reported high particle concentration during the winter season. A study by Kumar et al., 2018 looked at the seasonal variation of size distribution in size segregated aerosols and found PM<sub>0.95</sub> as



the dominant fraction in PM<sub>10</sub> concentration. Another recent study by Gani et al. (2020) has reported the long term measurements of aerosol size distribution over the megacity Delhi. They suggest that accumulation mode particle contributes the most to the PM<sub>2.5</sub> mass concentration over Delhi. Our result is in consistent with the above studies with high concentration of Aitken mode particle during all the four seasons of the Delhi with increase in accumulation mode particles during winter season.

## NPF Events: Observations and Characteristics

As discussed in the previous section, diurnal variation of aerosol particles showed an increase in nucleation mode particles during midday and warranted for further investigation. Hence, contour plots of aerosol size distribution have been done. Sudden burst of nucleation mode particles, revealing a banana type of growth were noticed from the contour plots. Further analyzing the mode diameter during the event shows a sudden dip towards the nucleation mode confirming the initiation of New Particle Formation (NPF). Contour plots of selected NPF event days of

different seasons are shown in **Figure 5** along with their diurnal variations in the mode diameter and mode wise number concentration. It shows that NPF events are generally observed at midday during the winter, summer and monsoon periods. A clear sudden burst of nucleation mode particles for a period of few hours is visible at all these days pointing to the NPF events (Dal Maso et al., 2005). The NPF starts at around 10:00 LT in the morning and continue to grow till 14:00 LT. Associated increase in number concentration on nucleation mode can also be seen in **Figure 5**. Aitken and nucleation mode are the dominant modes over the sampling site during the event days except for winter. The NPF event is preceded by an elevated Aitken mode particle concentration which drops as the NPF commences. During the NPF event time, N<sub>5-30</sub> shows a peak growth. In the representative figures for NPF events highest increase in nucleation mode number concentration events are seen during summer, which reaches  $\sim 3.1 \times 10^4 \text{ cm}^{-3}$ , following which the particle concentration decreases and continues to grow in Aitken mode which reaches a maximum concentration of  $\sim 4.5 \times 10^4 \text{ cm}^{-3}$ .

From a total of 222 days of data, 17 (8.4%) NPF event days have been identified along with 28 ( $\sim 12.4\%$ ) no NPF events days.

**TABLE 2** | Summary of the NPF events for different seasons at Delhi.

Date	Time period	GR <sub>nuc</sub>	CS <sub>avg</sub>
		(nm/h)	(10 <sup>-3</sup> s <sup>-1</sup> )
Summer			
31-03-2012	9:00–17:00	1.8	6.59 ± 2.68
03-04-2012	10:45–18:00	4.7	6.38 ± 3.83
20-04-2012	10:30–13:30	6.8	5.73 ± 7.93
21-04-2012	8:00–12:15	3.7	3.31 ± 1.53
24-04-2012	9:30–14:00	18.46	4.23 ± 3.25
27-04-2012	11:00–16:30	13.6	2.25 ± 1.11
06-06-2012	9:40–12:30	21.66	36.09 ± 20.39
11-06-2012	9:35–12:45	5.4	4.03 ± 6.74
13-06-2012	8:30–11:30	10.2	11.12 ± 4.14
18-06-2012	10:20–12:30	12.9	6.13 ± 4.27
19-06-2012	9:15–15:00	7.8	15.23 ± 6.25
28-06-2012	9:30–14:30	2.3	2.03 ± 1.04
30-06-2012	9:05–10:50	12.06	15.5 ± 33.3
		8.77 ± 6.25	9.31 ± 8.82
Monsoon			
04-07-2012	7:40–14:35	4.8	4.93 ± 2.60
16-07-2012	10:00–18:00	5.7	6.07 ± 2.29
		5.25 ± 0.64	5.5 ± 0.81
Winter			
13-02-2012	12:35–16:00	4.39	3.49 ± 1.13
16-02-2012	11:15–13:45	3.91	1.91 ± 0.73
		4.15 ± 0.34	2.65 ± 1.04

177 unidentified events have also been noted which corresponds to ~78.2% of the total data. Unidentified events are those days in which high level of background noise are present making it difficult to comprehend the features of number-size distribution in the data. This background noise could be due to the pollution or haze condition in the atmosphere. Also, the days on which if there were any doubts regarding the identification of clear NPF events then such days have been classified as unidentified days. An important point to be noted here is that there might be hidden NPF events in this unidentified event days however, the background noise hinders it from quantifying the particle growth. Among the 17 NPF events, 13 occurred in summer season, 2 in winter season and 2 during monsoon season. No events were observed in the post monsoon season. All the NPF event days showed a banana type of growth that continued for several hours and matched the previously reported NPF events in literature (Junninen et al., 2008; Lee et al., 2008). To understand the characteristics of new particle formation during all the seasons, the growth rate and condensation sink parameters have been studied.

**Table 2** summarizes the time period of the nucleation event, growth rate and condensation sink of the particle for different seasons. Almost all events start at much lower range in nucleation mode (<20 nm). Growth Rate for nucleation mode (GR<sub>nuc</sub>) particle is determined for the event days. In summer it ranges from 1.8 nm/h to 21.66 nm/h with a mean of 8.77 ± 6.25 nm/h. On a few occasions, a comparatively high GR<sub>nuc</sub> values were observed during the summer. Such high values of GR indicate the lesser time for the growth of nucleation mode particles. In monsoon GR<sub>nuc</sub> is noted to be about 4.8 nm/h and 5.7 nm/h with a mean of 5.25 ± 0.64 nm/h for the two events. Two daytime

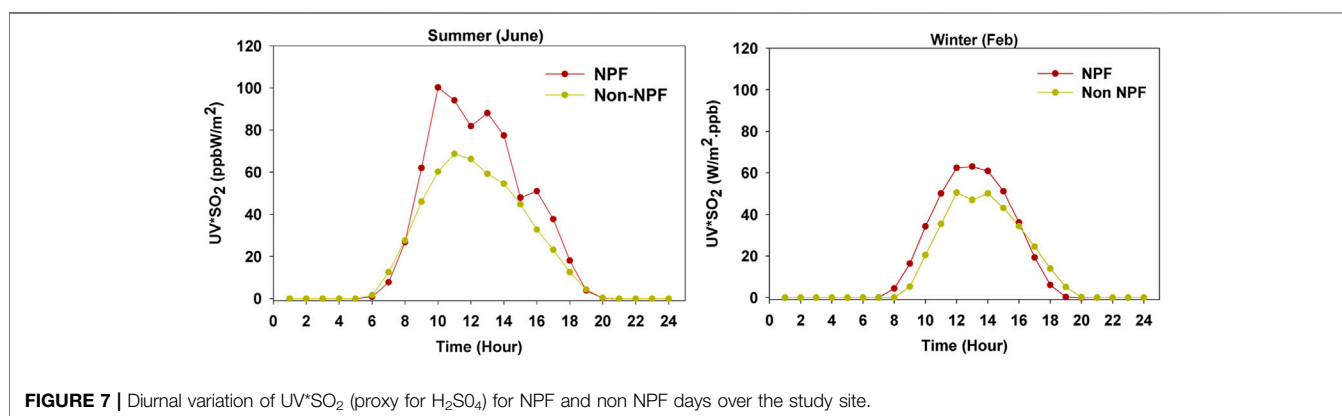
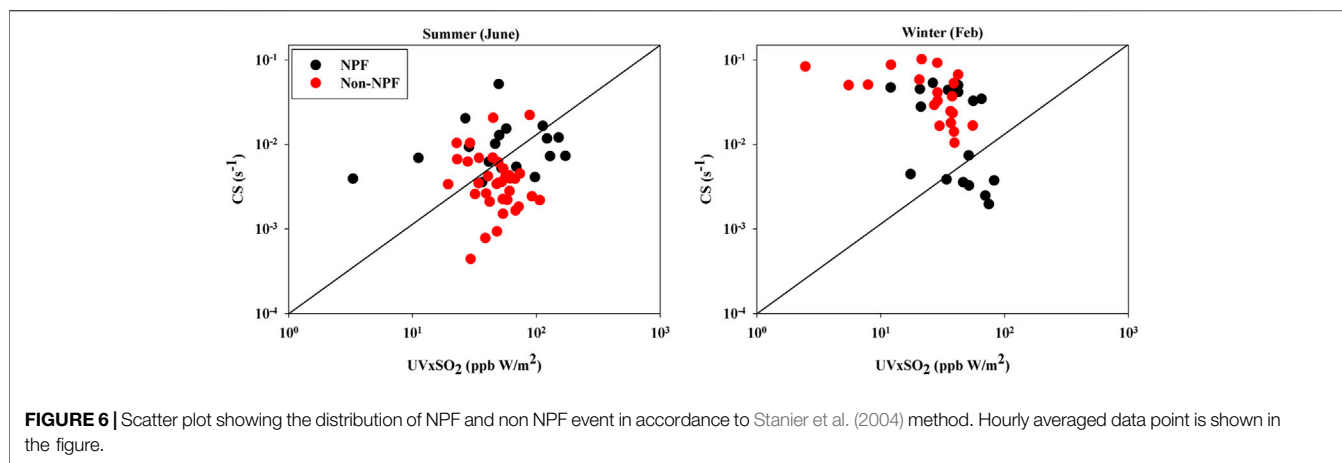
NPF events were observed during winter period whose GR<sub>nuc</sub> were 4.4 nm/h and 3.91 nm/h respectively. The mean GR<sub>nuc</sub> for summer and monsoon are comparatively higher than winter nucleation rates. Further, the GR<sub>nuc</sub> values of the present study at Delhi are slightly higher when compared with the regions such as Idaho hill (Weber et al., 1997); Ontario (Verheggen and Mozurkewich, 2002); Germany (Birmili et al., 2003) and Hyytiala, Finland (Dal Maso et al., 2007) but it is quite comparable with the GR values at most of the Indian locations reported, like Pune (Kamra et al., 2015); Kanpur (Kanawade et al., 2014) and Hanle, (Kompalli et al., 2014). At the coastal location of Trivandrum, however, the GR values are reported to be high (Kompalli et al., 2014) than that at Delhi.

## Discussion

Being a very polluted environment, the frequency of NPF events is notably less in Delhi. Out of 222 days, only 17 NPF events have been observed with maximum events (73.6%) occurring during the summer months and no NPF events reported during the post monsoon season. A number of factors influence the occurrence of NPF events at regional level including the abundance of precursor gases, pre-existing particle, solar radiation and the local meteorological conditions (Kerminen et al., 2018).

One of the influential factors for local NPF event is the amount of ambient gaseous H<sub>2</sub>SO<sub>4</sub> concentration, owing to its inherent connection to the gas phase chemistry associated with aerosol nucleation and subsequent growth (Lee et al., 2019). The solar radiation also acts as a driver for the new particle formation as it controls the formation of OH radicals and the concentration of H<sub>2</sub>SO<sub>4</sub> in the air (Pirjola et al., 1998; Dada et al., 2017). A decrease in the intensity of UV radiation tends to decrease the nucleation and the NPF events (Hamed et al., 2011; Baranizadeh et al., 2014). Stanier et al. (2004) used data of UVxSO<sub>2</sub> as a proxy to H<sub>2</sub>SO<sub>4</sub> and correlated it with the condensation sink (CS) data to see the conditions that favor nucleation. He found that for the same value of UVxSO<sub>2</sub>, nucleation took place at higher CS value during summer.

To analyse the possible mechanism for NPF events over the study region, we have studied different governing factors conducive for NPF events at selected dates from June 12, 2012 to June 18, 2012 for summer and February 13, 2012 to February 17, 2012 for winter. The time period is chosen in such a way that at least two NPF events are present for both the seasons and in accordance with the availability of SO<sub>2</sub> data. Monsoon events are not studied here, due to the unavailability of SO<sub>2</sub> data. As H<sub>2</sub>SO<sub>4</sub> data is unavailable for the study site, product of SO<sub>2</sub> and UV is used as a proxy to understand the chemistry on event and non-event days. In the present study hourly data of UVxSO<sub>2</sub> and condensation sink (CS) is correlated for event and nearby non-event days and plotted for the time window of 8:00 LT to 16:00 LT, keeping in view the occurrence of NPF events (**Figure 6**). It may be noticed that high CS and high UVxSO<sub>2</sub> is observed for NPF event day in summer and low CS and high UVxSO<sub>2</sub> was favorable for the NPF events in winter. We have also found that for the similar UVxSO<sub>2</sub> value nucleation takes place at higher CS values during summer as compared to winter. Unlike Stanier et al. (2004), where nucleation is usually more prominent on the right



side of the diagonal, our study do not show such clear preferences, possibly due to the presences of other predictive variables such as relative humidity, concentration of pre-existing particles and prevailing meteorological conditions.

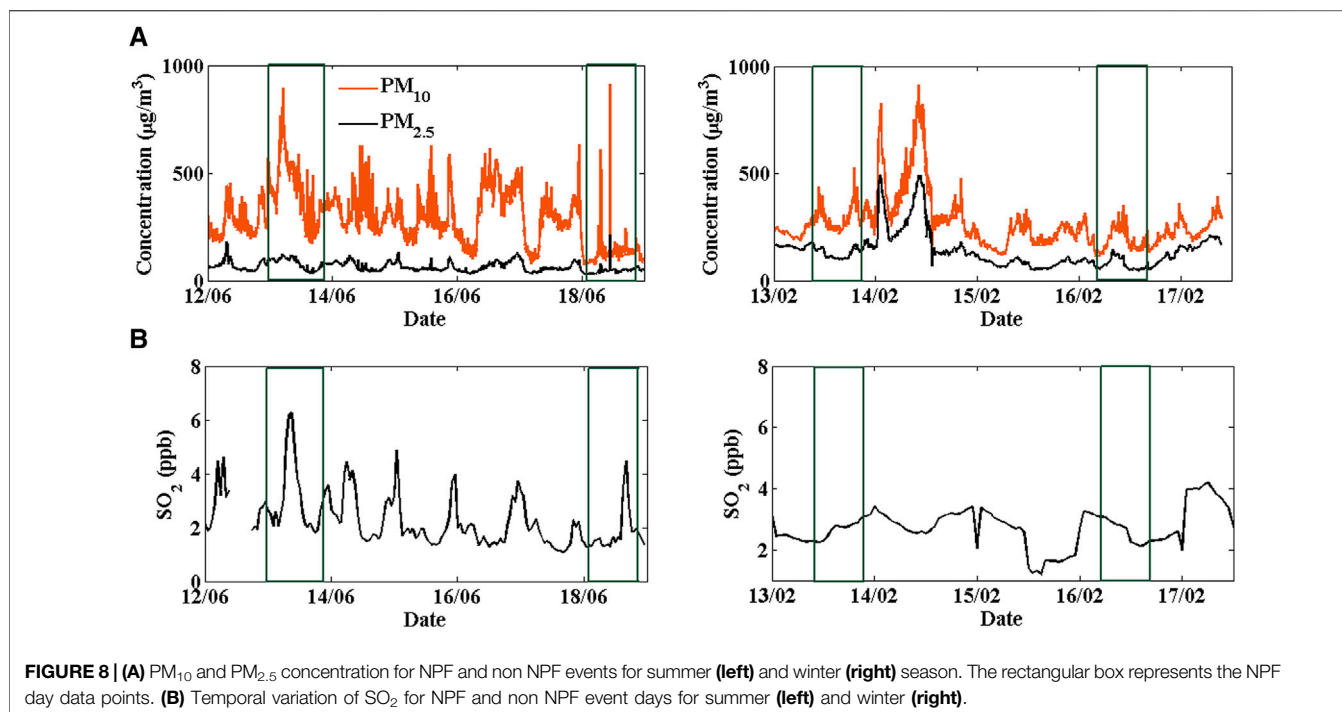
Over our study site, the amount of incoming solar radiation (both in shortwave and UV range) varies significantly during the summer and winter seasons. In summers, the day-time (7 AM–5 PM) shortwave and UV flux increase by 52 and 61% respectively, from the corresponding winter values. Thus, the abundant solar insolation during summer season (high UV) makes summer months more conducive for new particle formation, and this could be one of the plausible reasons for the increased NPF events during summer season. We have also looked at the diurnal variation of UVxSO<sub>2</sub> for event days and non-event days as plotted in **Figure 7**. It shows the diurnal variation in UVxSO<sub>2</sub> for June (summer) and February (winter). Considerable increase in UVxSO<sub>2</sub> is noticed during summer NPF days compared to the non-event days in summer. A similar result is seen for winter NPF event days also. However, the difference between NPF event and non-event days is much less during winter month. This also suggests that the abundant UV radiation along with high concentration of SO<sub>2</sub> (**Figure 8**) during the summer months could be the reason for the increased NPF event over the study region. A recent study by Jokinen et al. (2017) revealed the significance of photochemistry in NPF by studying the NPF

event during a partial solar eclipse day. He demonstrated the decrease in H<sub>2</sub>SO<sub>4</sub> due to decrease in UV during the eclipse time resulting in decreased nucleation events. Thus, the strong gas phase chemistry owing to the high UV radiation coupled with abundant precursor gas concentration (SO<sub>2</sub>) could be the reason for the high NPF events reported during the summer period over the study site.

Some of the previous studies have also shown that RH is negatively correlated with continental NPF events (Hamed et al., 2011; Dada et al., 2017). Increase in condensation and coagulation sink have been observed generally during high RH days, owing to the hygroscopic growth by pre-existing aerosol particles due to high intake of water vapor, leading to larger loss of condensable vapor such as H<sub>2</sub>SO<sub>4</sub> (Hamed et al., 2011). This may lead to much smaller nucleation rates or nucleation events. In our study, however, we have found that most of the NPF event occurred at RH less than 40%. As the number of NPF events are also less in the present case and mostly observed below 40% RH, a correlation between NPF event and the RH could not be established here.

In order to see the distribution of preexisting particles over the study area we have looked at the PM<sub>10</sub>, PM<sub>2.5</sub> and SO<sub>2</sub> concentration over the study region. **Figure 8** shows the distribution of PM<sub>10</sub>, PM<sub>2.5</sub> and SO<sub>2</sub> over the selected time window for the NPF and non-NPF days. Comparatively less





PM<sub>10</sub> concentration is observed for NPF days than the non NPF event days for both seasons which is in accordance with previous literatures. However, high PM<sub>10</sub> concentration is noticed for the summer NPF event on June 13, 2012 reaching up to 900  $\mu\text{g}/\text{m}^3$ . SO<sub>2</sub> concentration is also seen high on this day reaching to a concentration of 6 ppb. Delhi experiences high dust activity during summer months, could be a possible reason for the high PM<sub>10</sub> on the particular day. Compared to non-event days in summer, high CS and high PM<sub>10</sub> concentration have been noticed for NPF event days. This is a deviation from the usual NPF mechanism. However high CS along with high preexisting particle has also been noticed during NPF events in other regions as well (Yue et al., 2009; Nie et al., 2014). Study by Nie et al. (2014) suggest a possible mechanism for the dust induced NPF event, through dust induced heterogeneous photo catalytic reactions producing OH radicals enhancing the SO<sub>2</sub> production. Our study using proxy data for H<sub>2</sub>SO<sub>4</sub> suggests the strong gas phase chemistry owing to the high UV radiation coupled with abundant precursor gas concentration (SO<sub>2</sub>) could be the reason for the high NPF events reported during the summer period over the study site.

## CONCLUSION

Monthly and seasonal characteristics of the size distribution and number concentration of aerosol particles in the size range of 5 nm–32  $\mu\text{m}$  have been studied at Delhi using GRIMM-WRAS during December 2011 to January 2013 along with the simultaneous measurements of SO<sub>2</sub>, RH and solar radiation flux in the UV range. The findings of the study may be summarized as below:

The mean number concentration at Delhi during the entire experimental period is  $14.08 \pm 7.60 \times 10^3 \text{ cm}^{-3}$  with highest concentration during post-monsoon ( $15.8 \pm 4.26 \times 10^3 \text{ cm}^{-3}$ ) followed by winter ( $15.0 \pm 3.5 \times 10^3 \text{ cm}^{-3}$ ), monsoon ( $13.0 \pm 0.2 \times 10^3 \text{ cm}^{-3}$ ) and summer ( $10.5 \pm 5.5 \times 10^3 \text{ cm}^{-3}$ ). Study site is dominated by Aitken mode particle which follows a bimodal pattern in diurnal variation having peaks during morning and evening traffic hours. Accumulation mode particles are scarce over the study location except during the winter and post-monsoon days, when it shows a bimodal secondary peak along with the primary Aitken mode particle. The volume size distribution is also characterized by two modes, fine and coarse. Fine mode dominates during all the seasons except summer, when coarse mode dominates.

From 222 days of WRAS data, a total of 17 NPF events (13 in summer, 2 in winter and 2 during monsoon) have been observed. No NPF events were observed during post-monsoon season. During NPF events the Aitken and nucleation mode particles contribute more to the number concentration. The nucleation growth rate during summer and monsoon are more or less comparable and vary in the range of a few nm/h to ~20 nm/h, whereas, during winters it is much smaller in the range.

Our study on selected time period of NPF events show that high UV, low water vapor and low CS are favorable conditions for NPF events in winter. However, for summer high CS values are observed for NPF days, an aberration from previous studies, with increased PM<sub>10</sub> and SO<sub>2</sub> concentration in the background. This could be due to a possible NPF pathway through dust induced photo catalytic production of SO<sub>2</sub>. Proxy study using UVxSO<sub>2</sub> points to the strong influence of gas phase chemistry during summer months and hence the high frequency of NPF events. Myriads of factors contribute to NPF event. A detailed study of

precursor gases and VOCs are also required to reach conclusion on different mechanisms for NPF events on both seasons at the study site.

## DATA AVAILABILITY STATEMENT

The raw data supporting the conclusion of this article will be made available by the authors, without undue reservation.

## AUTHOR CONTRIBUTIONS

SJ: Execution, software and Programming, graphs and manuscript preparation. AM: Conceptualization, editing. NL:

Observation and Data. SSH: Observation and Data. SSI: Conceptualization, Supervision and editing.

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