Reviewer 1:

Comments:

We would like to thank the reviewer for their comments.

Delhi is well recognised as one of the world's most polluted megacities and is a city for which there are relatively few data available for highly resolved particle size distributions. This paper reports on around 15 months of continuous measurements of submicrometer particle size distributions and provides an interpretation which gives valuable insights into the processes which determine the size distribution. The length of the dataset also allows segregation according to seasons which is valuable. Calculations of the condensation and coagulation sink are a valuable complement to the data analysis process.

The work is of good quality but there are a number of points which, if addressed, would significantly enhance the value of the paper. These are as follows:

(a) Sampling took place on the campus of the Indian Institute of Technology in Delhi, but rather little detail is given of the sampling site apart from very local characteristics. The IIT Delhi campus is close to the southern perimeter of Delhi and some comment would be valuable on the degree to which this site could be considered as representative of Delhi more widely. Could we expect to see similar data from a site in the city centre or in Old Delhi? Were the prevailing winds from the city or from the less populated areas to the south?

Thank you for this comment and the opportunity to clarify this important point in the manuscript.

PN concentrations can be extremely variable even within a small spatial domain and definitely in a megacity like Delhi where two different sites can have unique types and strengths of sources emitting in different particle size ranges. While the main themes discussed in the manuscript (eg, rapid coagulation scavenging, suppression of UFP, etc) are most likely site independent given the large concentrations of fine PM across Delhi (and the Indo-Gangetic plain), the absolute PN concentrations can vary among sites in Delhi. Delhi has a prevailing North-western wind and would reach our site in South Delhi after traversing ~20 km of Delhi moving across industrial, commercial, and residential areas.

Added to Section 2.1 (Site details):

"The prevailing wind direction in Delhi is from the northwest, implying that the air we sampled would have traversed through ~20 km of industrial, commercial, and residential areas in Delhi before reaching our site."

Added to Section 2.4 (Limitations and uncertainties):

"Another limitation of this study is that our measurements were limited to a single site, and therefore do not capture the spatial variability within Delhi. UFP concentrations can be quite variable even within a small spatial domain (Saha et al., 2019; Puustinen et al., 2007), so it is

likely that a megacity like Delhi with diverse local sources will have strong spatially variability in PSD and PN concentrations. Future studies can quantify this spatial variability by measuring PSD at multiple fixed sites and using techniques such as mobile monitoring (e.g., Apte et al., 2017)."

(b) There are a number of questions concerning the instrumentation. Measurements were made with a TSI Scanning Mobility Particle Sizer. These are generally recommended to be used with an online dryer. Was any conditioning of the inlet air carried out, or do the measurements refer to ambient temperatures and relative humidity? Were the conditions inside the building where the instrument was located the same as those outdoors? Details are given of corrections applied for sampling losses. The TSI instrument software normally calculates sampling losses on the basis of detail of the instrument setup, and if this were carried out, there is a risk of applying two sets of corrections. Was the TSI correction turned off? Which version of the AIM software was used?

We have added some more information on setup and software to clarify these very relevant questions. Even more details of the setup can be found in Gani et al. (2019).

The sampled air passed through a $PM_{2.5}$ inlet and a Nafion membrane diffusion dryer. The room in which the instruments were installed was air conditioned and generally regulated to ~20–25°C. We used TSI's AIM version 9 software and sampling loss corrections were turned off as we applied our own corrections (discussed in manuscript).

We have included the following text in Sec. 2.2 (Instrumentation and setup) and Sec 2.3 (Data processing and analysis):

"The inlet had a $PM_{2.5}$ cyclone, followed by a water trap and a Nafion membrane diffusion dryer (Magee Scientific sample stream dryer, Berkeley, CA)."

"We used the Aerosol Instrument Manager version 9.0 (TSI, Shoreview, MN) software for logging data from the SMPS (software sampling loss corrections turned off)."

(c) The condensation sink was calculated using the properties of sulphur dioxide. Condensation sinks are more often calculated for sulphuric acid vapour and it is not at all clear why SO2 was assumed as the condensing vapour. Table 2 lists condensation and coagulation sinks from the literature referring to studies by Laakso et al. (2006) and Hussein et al. (2004). A search of these papers for the condensing molecule revealed no data for condensation sink or coagulation sink in either paper. This requires further explanation.

We recalculated the condensation sink for H_2SO_4 vapor and updated the values in Table 2 accordingly. Since the updated values calculated for H_2SO_4 are only different by ~10% than those calculated earlier for SO_2, the discussion remains unchanged.

We used the particle size distribution data from literature to calculate the condensation and coagulation sinks based on these PSDs. The specific PSDs used for these calculations for Delhi and Beijing can be found in Fig. 2 of Laakso et al. (2006) and for Helsinki in Table 1 of Hussein et al. (2004). We have updated the caption and footnotes in Table 2 to reflect these clarifications

and to include the specific figure/table from where we obtained the PSD used for the coagulation and condensation calculations.

Updates to Table 2:

Condensation sink values calculated for H_2SO_4 instead of SO_2.

Caption: "Characteristic coagulation and condensation sinks for Delhi and comparison with those calculated using PSD in literature from other cities."

Footnotes: "[a]Average PSD for Winter 8–9PM. [b]Average PSD for Monsoon 2–3PM. [c]PSD from Fig.2 in Laakso et al. (2006). [d]PSD from Table 1 in Hussein et al. (2004)."

(d) The discussion of particle number and mass concentrations and of size-resolved particle concentrations on pages 5-7 does not appear to account for restrictions applying in Delhi for the access of heavy duty vehicles which can only enter certain areas at nighttime. This seems likely to affect the particle number concentrations and size distributions and should be discussed. A further point which is wholly lacking from this part of the discussion is the relative contributions of locally-generated and advected particles. Many workers have highlighted the significance of biomass burning in the rural areas outside of Delhi and there are also highly polluting brick kilns. Can the contributions of such sources be identified? Does the airport (which is quite close) influence the measured data?

We have now included some more information on traffic volumes (total and truck traffic) in the manuscript (please refer to response to comment (f)). We agree that biomass burning (both inside and outside Delhi), brick kilns, possibly airport (4km West of our site), and other primary and secondary sources can contribute to number concentrations. While a detailed source apportionment of particle number concentrations is beyond the scope of this manuscript, our previous papers look at source apportionment for aerosol mass (Bhandari et al., 2020; Gani et al., 2019).

(e) On page 7 there is some discussion of comparison of concentrations with other cities but the literature cited is at least 10 years old. Particle number concentrations can change quite massively, as they have done in western Europe as a result of controls on the sulphur content of fuels. Hence, a comparison with such old data is probably of little value and this needs to be updated.

We have updated our literature review to include more recent papers.

"The N_{ait} peaks during the morning and the evening traffic rush hours are consistent with vehicular emissions contributing in this size range (Paasonen et al., 2016; Kumar et al., 2010). Studies from other cities—especially from near-roadway sites—also observe large fraction of the Aitken mode particles in the total PN concentrations (Zhu et al., 2002; Wu et al., 2008). In addition to traffic, other sources contributing to Aitken mode particles include cooking, industrial activities, solid waste burning, and construction activities (Kumar et al., 2013; Riffault et al., 2015; Chen et al., 2017)."

"In addition to emitting in the Aitken mode, biomass burning and older diesel vehicles emit in the accumulation mode as well (Chen et al., 2017; Paasonen et al., 2016; Kumar et al., 2010). Additionally, particles from fresh vehicular emissions can grow rapidly from the Aitken mode at the tailpipe to accumulation mode at roadside and ambient locations (Ning et al., 2013). We speculate that the traffic-related particles in Delhi may get smaller as India's vehicle fleet (especially heavy-duty trucks) is upgraded (Guttikunda and Mohan, 2014; Zhou et al., 2020a)."

(f) The section on multi-modal PSD fitting draws conclusions based upon assumed patterns of traffic. This would be strengthened by inclusion of a graph showing traffic volumes as a function of time of day, preferably for different vehicle classes.

Mishra et al. (2019) reported the traffic conditions at multiple sites in Delhi before and during the period when the odd-even driving scheme (January, 2016). At almost all sites, the morning rush hour was during 8–11am and the evening peak during 5–9pm. Furthermore, there was an increase in heavy duty truck traffic after 10pm (restriction of entry of trucks into Delhi relaxed).

"Delhi traffic usually peaks around 8–11am in the morning and 5–9pm in the evening. Restrictions on daytime heavy duty truck traffic have resulted in increased nocturnal truck traffic after 10pm (Mishra et al., 2019; Guttikunda and Calori, 2013).

(g) The comparator used for particle number concentrations in the conclusions section is the review of Kumar et al. (2014). It should be noted that many of the studies included in that review are now very old, and in line with the comment above, it would be more appropriate to compare with more recently collected data.

We have now generally updated the literature cited throughout the paper. We have removed the 'general PN range' from the conclusions since that would be influenced significantly by the instrumentation size limits (discussed in Sec. 2.4). Instead we have edited/included the following:

"While Delhi experiences PM levels 10 times higher than cities in North America and Western Europe, PN levels were comparable to those observed in urban sites in relatively cleaner (in terms of aerosol mass) cities."

The manuscript is well written and pleasingly clear of minor typographic errors. However, in Table 1 the units for PM0.56 are incorrect.

Typo fixed. Thank you for pointing this out. We have reviewed and updated the manuscript for this and some other typographic errors as well.

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Reviewer 2:

Comments:

This manuscript presents a thorough analysis on the behaviour of particle number size distribution, including its connection to the particle mass concentration and associated particle sources and atmospheric processes, in Delhi, India. The topic is important and the paper can be considered original enough to warrant its publication in Atmospheric Chemistry and Physics. The analysis is based on more a year of measurements, allowing the investigation to cover all the relevant seasons. The paper is scientifically sound and well written. There are, however, a few places where interpretation/discussion could be either broadened or improved a bit. My detailed comments in this regard are given below.

The authors explain in detail how they choose the particle density in determining particle mass concentrations from measured particle number size distributions. The total mass concentration is restricted to particles smaller than 560 nm in diameter (PM0.56). Do the authors have any data to tell how well PM0.46 reflects fine particulate matter or PM10 in Delhi? This would be useful information as most mass concentrations discussed in the scientific literature are based on PM2.5 or PM10, or both.

We have now included a comparison of SMPS-$PM_{0.56}$ and $PM_{2.5}$ to the supplement (Fig. S14). Slope = 0.60 and R^2 = 0.60 for the least square fit between SMPS-$PM_{0.56}$ and $PM_{2.5}$. The $PM_{2.5}$ data used for this comparison was based on a regulatory site 3 km from our site. We have also included a comparison of the C-PM_1 (Composition-PM_1 = NR-PM_1 + BC) and SMPS-PM_1 in Fig. S14 (Slope = 0.95 and R^2 = 0.83). SMPS-PM_1 was calculated by extrapolating the PSD to 1μm using PSD model fitting and C-PM_1 was based on data from an ACSM and Aethalometer at our site (Gani et al., 2019).

In talking about sub-3 nm particles (page 4, lines 24-28), the authors could include the recent overview by Kontkanen et al (2007, Atmos. Chem. Phys. 17, 2163-87) covering a number of field observations in different types of atmospheric environments.

Included citation for Kontkanen et al. (2017) as it is indeed pertinent to the discussion.

An interesting feature mentioned at the end of section 3.1 is that ultrafine particle mass concentration in Delhi are comparable to typical PM2.5 mass concentrations in measured in North America. In this context, it would be equally interesting to know how much higher ultrafine particle mass concentrations are in Delhi compared to those in cleaner urban environments in North America, Europe or Asia (for the latter, see. e.g. Bruggemann et al. 2009 Atmos. Environ. 43, 2456-63; Cheung et al. 2016 Atmos. Chem. Phys. 16, 1317-30; Xue et al. 2019 Environ Sci. Technol. 53, 39-49).

We have now included these studies with the following text:

"Overall the mass contributions of UFP ranged between 2.1 and 10.3 $\mu g\ m^{-3}$ depending on season and time of day. For contrast, mass concentrations of UFP have been observed to be <3 $\mu g\ m^{-3}$ in Germany (Brüggemann et al., 2009), <2 $\mu g\ m^{-3}$ in Taiwan (Cheung et al., 2016), and <0.2 $\mu g\ m^{-3}$ in California (Xue et al., 2019)."

When discussing how different factors (insolation, particle loading, RH) influence nucleation mode particles originating from atmospheric new particle formation (lines 12-18 on page 6 and lines 21-22 on page 10), a lot of new information has been obtained since the papers by Kulmala et al (2004) and Hamed et al. (2011), as reviewed in Kerminen et al. (2018, Environ. Res. Lett. 13, 102003). For example, the low particle mass concentrations and high insolation in summer act together to favor nucleation and survival of nucleated particles. Also, while a lower relative humidity seems to favor atmospheric nucleation, the actual reason for this observation has not been fully resolved yet.

We have now included this recent review paper along with following updates to the text in both sections mentioned:

 "The low particle mass concentrations and high insolation during the daytime of warmer months act together to favor nucleation and survival of nucleated particles (Kerminen et al., 2018), resulting in higher N_{nuc} concentrations during these periods."

"In addition to large condensation sinks from existing particles (Sec. 3.4), it is possible that the elevated RH during the cooler winter and autumn (Fig. 1) could further suppress atmospheric nucleation. It has been observed in both clean and polluted environments that the RH during new particle formation event days tends to be lower compared to non-event days (Kerminen et al., 2018; Dada et al., 2017; Hamed et al., 2011).

The authors mention specific dominating size ranges when discussing particles emitted from different sources (lines 27-32 on page 6, lines 5-7 on page 7 and lines 21-25 on page 11). Most of the references used in this discussion are 10-20 years old, and may not reflect the current state of knowledge or situation. For examples changes in vehicle engine technology over time or differences the general character of emission sources between India and both Europe and North America probably influences all this. Please check out the discussed "facts" based on more recent literature (e.g. Kumar et al. 2010 Atmos. Environ. 44, 5035-52; Kumar et al. 2013 Atmos. Environ. 67, 252-277; Riffault et al. 2015 Critical Reviews in Environmental Science and Technology 45, 2305-2356; Paasonen et al. 2016 Atmos. Chem. Phys. 16, 6823-40; Zhou et al. 2020 Atmos. Chem. Phys. 20, 1701-1722).

We have updated the references to incorporate suggestions from both reviewers and a few more including those specifically suggested in this comment (Kumar et al., 2010; Kumar et al., 2013; Riffault et al., 2015; Paasonen et al., 2016; Zhou et al., 2020).

The edits to the manuscript in the order mentioned in the comment by the reviewer are as follows:

"The N_{ait} peaks during the morning and the evening traffic rush hours are consistent with vehicular emissions contributing in this size range (Paasonen et al., 2016; Kumar et al., 2010). Studies from other cities—especially from near-roadway sites—also observe large fraction of the Aitken mode particles in the total PN concentrations (Zhu et al., 2002; Wu et al., 2008). In addition to traffic, other sources contributing to Aitken mode particles include cooking, industrial activities, solid waste burning, and construction activities (Kumar et al., 2013; Riffault et al., 2015; Chen et al., 2017)."

"In addition to emitting in the Aitken mode, biomass burning and older diesel vehicles emit in the accumulation mode as well (Chen et al., 2017; Paasonen et al., 2016; Kumar et al., 2010). Additionally, particles from fresh vehicular emissions can grow rapidly from the Aitken mode at the tailpipe to accumulation mode at roadside and ambient locations (Ning et al., 2013). We speculate that the traffic-related particles in Delhi may get smaller as India's vehicle fleet (especially heavy-duty trucks) is upgraded (Guttikunda and Mohan, 2014; Zhou et al., 2020a)."

"The relatively smaller median diameter for the accumulation mode during the traffic rush hour is consistent with particles from fresh vehicle exhaust—both gasoline and diesel—being smaller than those from biomass burning and other aged aerosol. While gasoline engines emit in the ~20–60 nm range, some of the older diesel engines still prevalent in India can emit larger accumulation mode particles (Kumar et al., 2010). Particles from biomass burning are in the ~50–200 nm size range (Chen et al., 2017; Janhäll et al., 2010). For the warmer periods which have relatively less biomass burning emissions, the ultrafine modes contribute to relatively higher fraction of PN—especially during traffic rush hours."

I appreciate the analysis on coagulation scavenging of nucleation mode particles (section 3.3). There are a couple of issues worth to be discussed in more detail here. First, since the lifetime of a nucleation mode particles is sensitive to their diameter, it is essentially a competition between particle growth and scavenging by coagulation that matter the most, not only the lifetime itself (Pierce and Adams 2007 Atmos. Chem. Phys. 7, 1367-79; Lehtinen et al. 2007 J. Aerosol Sci. 38, 988-994). This competition becomes even more important when approaching the sizes at which particles are nucleation, which brings us to the second point: how is it possible that nucleation takes place at all in polluted megacities? This question was investigated by Kulmala et al. (2017, Faraday Discuss. 200, 271-288) but no definite answer could be pointed out. This unsolved issue could be mentioned briefly in this context. Third, there is one more paper related to this topic (Cai and Jiang 2017, Atmos. Chem. Phys. 17, 12659-75) that could be considered here and also when first bringing up the important role of coagulation scavenging ín determining the fate of ultrafine particles (page 2, line 25).

We agree with the reviewer about the competition between growth rate and the coagulation scavenging. We calculated the survival parameter defined in Kulmala et al. (2017) and found that even for the least polluted period in Delhi we should not be able to observe NPF (P>50 for the least polluted period). This finding was consistent with the discussion on megacities in China in Kulmala et al. (2017). We have included the following text in our discussion on NPF (end of Section 3.4):

"Based on the survival parameter ($P = (CS/10^4 \text{ s}^{-1})/(GR/\text{nm h}^{-1})$) defined in Kulmala et al. (2017), new particle formation would not be expected even during some of the least polluted periods in Delhi. Why we observe any new particle formation events in Delhi—similar to observations from megacities in China—is still not well understood (Kulmala et al., 2017)."

We have also added Cai and Jiang (2017) to the discussion on coagulation scavenging in determining the fate of UFP in China (Section 1).

The discussion about the particle growth rates GR (page 9, lines 17-19) need to be revised/updated. First, there are clear differences in typical values (and value ranges) of GR between different types environments, and second, particle growth rates in polluted megacities do not seem to be usually below 5 nm/hour (Nieminen et al. 2018 Atmos. Chem. Phys. 18, 14737-56; Kerminen et al. 2008 Environ. Res. Lett. 13, 102003; Chu et al. 2019 Atmos. Chem. Phys. 19, 115-138).

We agree with the reviewer. We have revised this discussion based on latest literature. We have also rephrased our discussion around the absence of new particle formation and growth during the polluted periods observed in Delhi.

"The growth rate of the nucleation mode particles based on the average PSD for these clean conditions was ~5 nm h^{-1}. It should be noted that our SMPS measurements had a lower size cut-off of 12 nm, implying the particles in the nucleation mode had already grown before being detected.

Growth rates depend on particle size, concentrations of condensable gases (H_2SO_4, highly oxygenated molecules, etc), and condensation and coagulation sinks, and they can range from 0.1 to 10s of nm h^{-1} across various environments (Kerminen et al., 2018; Nieminen et al., 2018; Bianchi et al., 2016). We did not observe banana-shaped growth patterns during the polluted winter or autumn seasons as the growth phase was probably disrupted by coagulation scavenging. For example, during winter, coagulation timescales for a 15 nm particle were <1 h (Fig. 8). Growth rates for polluted megacities also usually slower than 10 nm h−1 (Zhou et al., 2020b; Chu et al., 2019). It is likely that a nucleation mode particle is more likely to coagulate with a larger particle than to grow to the Aitken/accumulation mode. Overall, the aerosol dynamics are a complex interplay of both new particle formation and coagulation scavenging, with nucleation mode particles in highly polluted environments generally susceptible to scavenging by the accumulation mode particles."

I do not understand the last statement in section 3.5 (page 10, lines 22-24). Please explain more clearly and justify.

We have now added some more context to this concluding statement of that section.

Meteorology, specifically changes in the ventilation coefficient, have a strong influence in the seasonal difference in aerosol mass loadings in Delhi. However, we do not observe this strong influence of meteorology on particle number concentrations. While warmer periods (eg, summer) have lower mass loadings, the lower condensation/coagulation sink along with the

higher insolation results in higher number concentrations. The cooler polluted periods (eg, winter) have higher aerosol mass loadings and consequently a larger condensation/coagulation sink, suppressing the number concentrations. So, while ventilation coefficient has an inverse relationship with the mass loadings (ventilation higher = lower PM concentrations), ventilation coefficient does not influence number concentrations in the same straightforward manner that it modulates mass concentrations.

The new concluding statements for Section 3.5:

"Overall, ventilation strongly influences aerosol mass loadings in Delhi, with higher ventilation often resulting in reduction of PM concentrations and vice versa. However, we do not observe such a simple relationship between ventilation and PN levels. The complex interplay of sources, aerosol dynamics, and atmospheric processes as discussed above make PN levels less sensitive to meteorological changes as compared to aerosol mass loadings."

Do the authors have a concrete explanation on why rain would affect much more accumulation mode particles than Aitken or nucleation mode particles (lines 9-11 on page 10)? Is that 1) because accumulation mode particles are scavenged more efficiently by rain compared with smaller particles (is that true at all?), or 2) because accumulation mode particle have overall longer atmospheric lifetimes compared with smaller particles (so that compared with smaller particles, it would take longer to build up an accumulation mode after any individual rain event)?

Based on our observations, there does not seem to be a clear reduction in UFP concentrations during the rain event (at least not to the degree observed for accumulation mode particles). The lower reduction of the Aitken mode particles seems to be consistent with parametrization studies (Pryor et al., 2016; Laakso et al., 2003). However, these studies suggest that the nucleation mode particles should be scavenged more than Aitken mode particles. It is possible that the rain washout of nucleation mode particles is compensated by the reduction in coagulation/condensation sink resulting from a reduction in the accumulation mode particles.

"Comparing rainy and non-rainy days during the monsoon, our observations suggest that rain reduces the accumulation mode concentrations (Fig. S12) as has been observed in other studies (Luan et al., 2019; Hussein et al., 2018). However, we do not observe a similar reduction in either nucleation or Aitken mode particle concentrations. Parametrization studies suggest that rain is more effective in scavenging nucleation mode particles as compared to Aitken mode particles (Pryor et al., 2016; Laakso et al., 2003). While we cannot fully reconcile our observations here with theory, possible explanations include (i) differential water solubility by particle size and age, or (ii) that the rain washout of the nucleation mode particles is 'balanced' by the reduction in the condensation and coagulation sink due to rain washout of accumulation mode particles."

As a minor technical remark, a correct way to express ranges of quantities is to write "range from M to N", "range between M and N" or "are in the range M-N". Such ranges are incorrectly written in a few places, please correct.

Thank you for bringing this point to our attention. We have updated the manuscript to make it consistent with this remark.

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Particle number concentrations and size distribution in a polluted megacity: The Delhi Aerosol Supersite study

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Abstract.

The Indian national capital, Delhi, routinely experiences some of the world's highest urban particulate matter concentrations. While fine particulate matter ($PM_{2.5}$) mass concentrations in Delhi are at least an order of magnitude higher than in many western cities, the particle number (PN) concentrations are not similarly elevated. Here we report on 1.25 years of highly time resolved particle size distributions (PSD) data in the size range of 12–560 nm. We observed that the large number of accumulation mode particles—that constitute most of the $PM_{2.5}$ mass—also contributed substantially to the PN concentrations. The ultrafine particles (UFP, D_p <100 nm) fraction of PN was higher during the traffic rush hours and for daytimes of warmer seasons—consistent with traffic and nucleation events being major sources of urban UFP. UFP concentrations were found to be relatively lower during periods with some of the highest mass concentrations. Calculations based on measured PSD and coagulation theory suggest UFP concentrations are suppressed by a rapid coagulation sink during polluted periods when large concentrations of particles in the accumulation mode result in high surface area concentrations. A smaller accumulation mode for warmer months results in increased UFP fraction, likely owing to a comparatively smaller coagulation sink. We also see evidence suggestive of nucleation which may also contribute to the increased UFP proportion during the warmer seasons. Even though coagulation does not affect mass concentrations, it can significantly govern PN levels with important health and policy implications. Implications of a strong accumulation mode coagulation sink for future air quality control efforts in Delhi are that a reduction in mass concentration, especially in winter, may not produce proportional reduction in PN concentrations. Strategies that only target accumulation mode particles (which constitute much of the fine $PM_{2.5}$ mass) may even lead to an increase in the UFP concentrations as the coagulation sink decreases.

1 Introduction

Outdoor air pollution has detrimental health effects (Pope and Dockery, 2006; Schraufnagel et al., 2019a) and is responsible for more than 4 million deaths every year globally (Cohen et al., 2017), resulting in substantial global and regional decrements in life expectancy (Apte et al., 2018). There has been a continued growth of megacities (population > 10 million) and as of 2018 there are 47 megacities (United Nations, 2018). Many of the lower-income megacities experience persistently severe air

pollution problems and Delhi (population = 28 million) routinely experiences the highest annual-average fine particle mass (PM$_{2.5}$) concentrations of any megacity in the world (World Health Organization, 2018). In addition to aerosol mass, there is now growing evidence of potential health risks associated with ultrafine particles (UFP, D$_p$<100 nm). These particles can penetrate and deposit deep within the lungs, may enter the bloodstream, and reach sensitive internal organs (Yeh and Schum, 1980; Oberdörster et al., 2005; Salma et al., 2015; Schraufnagel et al., 2019b). While UFPs scarcely contribute to aerosol mass (PM), they have been observed to dominate particle number (PN) concentrations observed in urban environments (Hussein et al., 2004; Rodríguez et al., 2007; Wu et al., 2008).

Atmospheric particles size distributions (PSD) are often categorized by the nucleation mode (3–25 nm), the Aitken mode (25–100 nm) and the accumulation mode (100–1000 nm). UFP include both the nucleation and the Aitken modes. Most of the PM$_{2.5}$ mass is usually contributed from the accumulation mode particles, whereas UFP constitute most of the PN (Seinfeld and Pandis, 2006). Given that different modes of the PSD contribute disproportionately to aerosol mass (accumulation mode for PM$_{2.5}$) and aerosol number (UFP for PN) concentrations, PN concentrations do not necessarily share the same spatial or temporal dynamics as PM (Johansson et al., 2007; Puustinen et al., 2007; Reche et al., 2011). The accumulation mode particles in urban environments result from a wide range of anthropogenic sources (traffic, industrial, biomass burning, etc.), natural sources (wildfires, volcanoes, deserts, etc.), and chemical processing of gases and aerosol (Robinson et al., 2007; Zhang et al., 2007; Jimenez et al., 2009; Calvo et al., 2013). However, traffic (Zhu et al., 2002; Charron and Harrison, 2003; Fruin et al., 2008; Wehner et al., 2009), cooking (Saha et al., 2019; Abernethy et al., 2013), and new particle formation (Brines et al., 2015; Hofman et al., 2016) are generally considered the only major sources of urban UFP.

In cities with high aerosol mass loadings (e.g., Beijing and Delhi), coagulation scavenging can be a major sink of UFP—the numerous but smaller UFP are 'lost' to the fewer but larger particles that contribute most to the aerosol mass (Kerminen et al., 2001; Zhang and Wexler, 2002; Kulmala, 2003; Kulmala and Kerminen, 2008). Specifically, coagulation plays a crucial role in rapid removal of freshly nucleated particles onto pre-existing larger particles (Kerminen et al., 2001; Kulmala, 2003). Short-term studies from Delhi have observed coagulation as a major sink for UFP (Mönkkönen et al., 2004a, 2005), similar to observations from polluted cities in the USA in the 1970s (Husar et al., 1972; Whitby et al., 1972; Willeke and Whitby, 1975) and contemporary polluted cities in China (Wu et al., 2008; Peng et al., 2014)(Cai and Jiang, 2017; Peng et al., 2014; Wu et al., 2008). Polluted megacities such as Delhi often experience aerosol mass concentrations that are an order of magnitude higher than those experienced in cities in high-income countries (Pant et al., 2015; Jaiprakash et al., 2017; Gani et al., 2019), yet PN concentrations are not high in similar proportions (Mönkkönen et al., 2004b; Apte et al., 2011).

As a part of the Delhi Aerosol Supersite (DAS) study we investigated Delhi's aerosol chemical composition and its sources (Gani et al., 2019; Bhandari et al., 2020a)(Gani et al., 2019; Bhandari et al., 2020b). Here we use long-term observations from Delhi to present seasonal and diurnal profiles of PSD in this polluted megacity. We also provide insights into the phenomena—such as coagulation—that influence PSD in Delhi and interpret their implications for future policy measures. This study has the potential to help understand processes that drive PSD in other cities with similar sources and meteorology across the Indo-Gangetic plain (Kumar et al., 2017; Singh et al., 2015) and is relevant for other highly polluted urban environments.

2 Methods

2.1 Sampling site

We installed a suite of high time resolution online aerosol measurement instrumentation at the Indian Institute of Technology
Delhi (IIT Delhi) campus in South Delhi. The instruments were located on the top floor of a 4-story building and the nearest source of local emissions is an arterial road located 150 m away from the building. Delhi experiences a wide range of meteorological conditions with large diurnal and seasonal variations in temperature, RH and wind speed, among other parameters (Fig. 1). Furthermore, shallow inversion layers also occur frequently, especially for cooler periods. The prevailing wind direction in Delhi is from the northwest, implying that the air we sampled would have traversed through ~20 km of industrial, commercial, and residential areas in Delhi before reaching our site. We discussed the meteorology of Delhi in further detail in an earlier publication (Gani et al., 2019).

2.2 Instrumentation and setup

We measured PSD using a scanning mobility particle sizer (SMPS, TSI, Shoreview, MN) consisting of an electrostatic classifier (TSI model 3080), differential mobility analyzer (DMA, TSI model 3081), X-ray aerosol neutralizer (TSI model 3088), and a water-based condensation particle counter (CPC, TSI model 3785). Chemical composition of non-refractory PM_1 (NR-PM_1) was obtained using an Aerodyne Aerosol Chemical Speciation Monitor (ACSM, Aerodyne Research, Billerica MA) and BC was measured using a multi-channel aethalometer (Magee Scientific Model AE33, Berkeley, CA). The inlet had a $PM_{2.5}$ cyclone, followed by a water trap and a Nafion membrane diffusion dryer (Magee Scientific sample stream dryer, Berkeley, CA). Further details of the instrumentation, sampling setup, and the data processing methodology for the instrumentation (SMPS, ACSM and aethalometer) and meteorological parameters are provided in Gani et al. (2019).

2.3 Data processing and analysis

The SMPS at our site scanned from 12 to 560 nm with each subsequent scan 135 seconds apart. We used the Aerosol Instrument Manager version 9.0 (TSI, Shoreview, MN) software for logging data from the SMPS (software sampling loss corrections turned off). We calculated the transmission efficiency for the DAS sampling line based on the diffusion (Ingham, 1975) and settling (Pich, 1972) losses. We correct the observed PSD for these transmission efficiency losses and calculate the number concentrations (individual size modes and total) and the median diameter based on the updated PSD. Multi-modal lognormal fitting has been widely used to parameterize the PSD (Whitby, 1978; Seinfeld and Pandis, 2006). The parameterization allows for a quantitative description of the PSD and easier comparisons between different PSD data sets. Separating the various PSD modes can also help understand role of sources in driving each mode. We use a multi-modal fitting technique described in Hussein et al. (2005) to estimate individual log-normal modes of the PSD (Sec. 3.6). We included PSD data from January-2017 to April-2018 in this study.

Studies from Asia have estimated aerosol densities ~~between~~ in the range 1.3–1.6 g cm^3 (Sarangi et al., 2016; Hu et al., 2012). A previous study from Delhi used a density of 1.7 g cm^{-3} to estimate aerosol mass from their PSD observations (Laakso et al., 2006). Based on aerosol composition data, we estimated a density of ~1.5 g cm^{-3} for the non-refractory portion of the aerosol. However, that did not include the BC and metal component of the aerosol mass (we did not measure metals). We assume an overall average particle density of 1.6 g cm^{-3} as it provides a better mass closure (Gani et al., 2019) and is within the uncertainty bounds based on previous density estimates as well. We use this assumed density to estimate the mass in the observed PSD (PM$_{0.56}$), ultrafine particles (PM$_{UFP}$) and accumulation mode particles (PM$_{acc}$).

We calculated the Fuchs form of the Brownian coagulation coefficient (K_{12}) using equations published by Seinfeld and Pandis (2006) and then estimated the characteristic coagulation timescale ($\tau_{coag,i}$) of a particle of any size onto particles of any size greater than that particle using the technique of Westerdahl et al. (2009). We calculate condensation sink (CS) for vapor condensing on the aerosol distribution following Kulmala et al. (2001, 1998) and the growth rate (GR) following Kulmala et al. (2012).

$$\tau_{coag,i} = \frac{1}{\sum\limits_{i<j} K_{i,j} N_j} \tag{1}$$

$$CS = 4\pi D \sum_i \beta r_{p,i} N_i \tag{2}$$

$$GR = \frac{\Delta D_p}{\Delta t} = \frac{D_{p2} - D_{p1}}{t_2 - t_1} \tag{3}$$

where $K_{i,j}$ is the coagulation coefficient for particle in bin i of the PSD coagulating with those in bin j and N_j is the concentration of particles in bin j of the PSD. D is diffusion coefficient of the condensing vapor (we use ~~SO$_2$~~ H$_2$SO$_4$ for our calculations), β is the transitional correction factor, $r_{p,i}$ is the radius of a particle in bin i of the PSD, and N_i is the concentration of particles in bin i of the PSD. D_{p1} and D_{p2} are the diameters of the particles at times t_1 and t_2, respectively.

For our analysis, we categorize the seasons as winters (December to mid-February), spring (mid-February to March), summers (April to June), monsoon (July to mid-September) and Autumn (mid-September to November) (Indian National Science Academy, 2018). We define day as 7 AM–7 PM and night as 7 PM–7 AM.

2.4 Limitations and uncertainties

Like most measurement studies, the limitations and the uncertainties of this study arise from instrumentation and the sampling system. While we have applied correction for diffusion and settling losses in our sampling system, future studies can reduce sampling losses and subsequent uncertainties—especially for ultrafine particles—by designing shorter sampling systems with higher flow rates. The SMPS used for the PSD measurements has a ~30% uncertainty for submicron aerosol (Buonanno et al., 2009). Furthermore, we only measure particles larger than 12 nm, potentially underestimating concentrations of nucleation

mode particles. Our study does not provide any information on nanocluster aerosol (1–3 nm) which can contribute to a significant fraction of PN concentrations and their detection made possible by recent advances in aerosol measurement instrumentation ~~(Rönkkö et al., 2017; Kangasluoma and Attoui, 2019)~~(Rönkkö et al., 2017; Kontkanen et al., 2017; Kangasluoma and Attoui, 2019). Measuring the complete nucleation mode range along with these nascent nanoclusters can advance our knowledge of particle

5   formation pathways and their subsequent growth that leads to high air pollution (Yao et al., 2018; Guo et al., 2014).

     Another limitation of this study is that our measurements were limited to a single site, and therefore do not capture the spatial variability within Delhi. UFP concentrations can be quite variable even within a small spatial domain (Saha et al., 2019; Puustinen et al., 200 so it is likely that a megacity like Delhi with diverse local sources will have strong spatially variability in PSD and PN concentrations. Future studies can quantify this spatial variability by measuring PSD at multiple fixed sites and using techniques

10   such as mobile monitoring (e.g., Apte et al. (2017)).

3   Results and discussion

3.1   Particle number and mass concentrations

Delhi experiences large seasonal and diurnal variations in aerosol mass concentrations. In Gani et al. (2019), we showed that the wintertime submicron aerosol concentration was ~2× higher than spring and ~4× higher than the warmer months. In Fig. 2

15   we compare the seasonal changes in aerosol mass and PN concentrations. In contrast to the sharp seasonal variation in aerosol mass loadings, PN levels had much lower variability. The average PN levels for winter were 52500 cm^{-3}, spring 49000 cm^{-3}, summer 43400 cm^{-3}, monsoon 35400 cm^{-3}, and autumn 38000 cm^{-3}. The differences in the variability of number and mass concentrations are potentially explained by the difference in the sources and processes that drive their concentrations (Sec. 1).

     For each season, PN concentrations varied by the time of day (Fig. 3). In winter, average hourly PN concentrations ranged

20   between 32400 cm^{-3} and 77500 cm^{-3}. The hourly averaged winter PN levels had a morning (08:00) peak, followed by lowest concentrations during the afternoon (14:00), and highest concentrations during the evening (21:00). The spring hourly averaged concentrations ranged between 38100 cm^{-3} and 62400 cm^{-3}. The hourly averaged spring levels also had the morning (08:00) and evening (21:00) peaks, and the PN concentrations were lowest around 15:00. Summer hourly averaged concentrations ranged between 35900 cm^{-3} and 54200 cm^{-3}. The hourly averaged summer levels also had the morning (08:00) and evening

25   (21:00) peaks, there was also a small peak during the midday (11:00) and the PN concentrations were lowest around 03:00. Monsoon hourly PN concentrations ranged between 25800 cm^{-3} and 43500 cm^{-3}. The monsoon PN hourly average concentrations were lowest during early morning (04:00), highest during the midday (12:00), followed by another evening peak in the evening (21:00). Autumn hourly average PN concentrations ranged between 25500 cm^{-3} and 49200 cm^{-3}. The autumn diurnal profile had a morning (07:00) and an evening peak (21:00), with the concentrations lowest during the daytime (13:00). While

30   hourly averaged PN concentrations for all seasons had peaks during the morning and the evening, additional midday peaks were observed during the summer and monsoon.

     In Fig. 3 we also compare the seasonal and diurnal changes in PN and $PM_{0.56}$ concentrations—number and mass concentrations observed from the SMPS. The SMPS-based mass concentrations follow the diurnal profile similar to those of aerosol

mass loadings in Delhi (Gani et al., 2019). However, PN diurnal profiles do not closely follow those of $PM_{0.56}$ (campaign R^2 of the hourly averaged time-series of PN and $PM_{0.56}$ = 0.26). The low correlation of aerosol number and mass is consistent with UFP making up a large fraction of the PN (Sec. 3.2), but contributing much less to the aerosol mass compared to larger accumulation mode particles. In Fig. 4, we estimate the aerosol mass loading in the ultrafine mode and the accumulation mode particles. While PM_{acc} generally accounted for more than 90% of the $PM_{0.56}$ concentrations, the PM_{UFP} fraction of $PM_{0.56}$ ranged between 3 —10and 10% depending on season and time of day. The fractional PM_{UFP} contribution to $PM_{0.56}$ was highest during the evening traffic rush hours for the summer and monsoon, and lowest during the non-traffic rush hours of the winter and autumn. The average wintertime PM_{UFP} concentrations ranged from 2.8 µg m^{-3} for midday to 10.3 µg m^{-3} during the evening traffic. For the other seasons, average PM_{UFP} concentrations were between in the range ~2–5 µg m^{-3}. Overall, the mass contributions of UFP were ranged between 2.1 and 10.3 µg m^{-3} depending on season and time of day. Even For contrast, mass concentrations of UFP have been observed to be <3 µg m^{-3} in Germany (Brüggemann et al., 2009), <2 µg m^{-3} in Taiwan (Cheung et al., 2016), and <0.2 µg m^{-3} in California (Xue et al., 2019). Furthermore, even if the fractional contribution to total mass in Delhi remains low (<10%), the absolute mass concentrations from the observed UFP during polluted periods were nearly as high as the $PM_{2.5}$ concentrations observed in many cities in North America (~8–12 µg m^{-3}) (Manning et al., 2018).

3.2   Size resolved particle concentrations

The PN concentrations and mass loadings are driven by the concentrations of the individual size modes—nucleation (N_{nuc}), Aitken (N_{ait}), and accumulation (N_{acc}). It should be noted that the N_{nuc}, N_{ait}, and N_{acc} refer to the specific particle size range and not the individual lognormal modes that constitute the PSD (Sec. 3.6). While the N_{nuc} and N_{ait} constitute the UFP concentrations (UFP = N_{nuc} + N_{ait}), N_{acc} contributes to both the total PN concentrations (PN = N_{nuc} + N_{ait} + N_{acc}) and most of the fine aerosol mass (Seinfeld and Pandis, 2006). In Fig. 3 we present the seasonal and diurnal variation of N_{nuc}, N_{ait}, N_{acc}, UFP, PN, and the median particle diameter. In Fig. 5 we present the stacked absolute and fractional diurnal profiles of the PN components for each season to illustrate the varying contribution of each size mode to the PN concentrations by season and time-of-day. Furthermore, in Table 1 we present the summary daytime and nighttime averages for all seasons.

The nucleation mode particle concentrations were highest (in magnitude and fraction of PN) during the warmer periods, when particle mass concentrations tend to be lower. The average hourly N_{nuc} peaked around 11:00–12:00 for spring, summer, and monsoon. These N_{nuc} peak hourly averaged concentrations were ~18000 cm^{-3} and contributed to almost 40–50% of the PN concentrations for these periods. For nighttime of spring, summer and monsoon and for all times of day for autumn and spring, hourly averaged N_{nuc} concentrations were usually less than 10000 cm^{-3} and contributed to within 10–20% of the PN concentrations. The increase in low particle mass concentrations and high insolation during the daytime of warmer months act together to favor nucleation and survival of nucleated particles (Kerminen et al., 2018), resulting in higher N_{nuc} concentrations during the daytime is consistent with new particle formation during high insolation periods(Kulmala et al., 2004)these periods. In Sec. 3.3, we discuss the strong condensation and coagulation sink during extremely polluted periods (such as winter and autumn) and its role in suppressing N_{nuc} concentrations.

The Aitken mode particle concentrations generally constituted the largest fraction of the PN concentrations. N_{ait} contributed to 40–60% of PN concentrations depending on season and time of day. N_{ait} concentrations were highest during the morning and evening hours—periods with lower ventilation and high vehicular traffic. The average hourly N_{ait} concentrations ranged between 12000 cm^{-3} (monsoon early morning) to 44300 cm^{-3} (winter evening). The hourly averaged N_{ait} profiles for most seasons had a morning and an evening peak. For winter, autumn, and spring, the hourly averaged N_{ait} concentrations were lowest during the midday. However, for summer and monsoon there was another peak during the midday with lowest levels observed early in the morning. The small daytime N_{ait} peaks for the warmer months are consistent with nucleation mode particles growing to the Aitken mode (Kulmala et al., 2004). The N_{ait} peaks during the morning and the evening traffic rush hours are consistent with vehicular emissions contributing in this size range ~~(Ristovski et al., 1998; Morawska et al., 1998)~~(Paasonen et al., 2016; Kumar et al., 2010). Studies from other cities—especially from near-roadway sites—also observe large fraction of the Aitken mode particles in the total PN concentrations (Zhu et al., 2002; Wu et al., 2008). In addition to traffic, ~~the elevated N_{ait} concentrations during the morning and evening could also be contributed from biomass burning which results in particles sizes ~50–200 nm (Chen et al., 2017)~~other sources contributing to Aitken mode particles include cooking, industrial activities, solid waste burning, and construction activities (Kumar et al., 2013; Riffault et al., 2015; Chen et al., 2017).

Accumulation mode particles contribute to almost all of the submicron aerosol mass, so it is unsurprising that the diurnal and seasonal variation of N_{acc} concentrations are similar to those of aerosol mass concentrations (Gani et al., 2019). In addition to contributing to most of the fine PM mass concentrations, N_{acc} in Delhi also constituted a significant fraction of the PN concentrations. The N_{acc} fraction of PN were the highest for nighttimes of autumn and winter (~45%) and the lowest for the daytimes of the warmer summer and monsoon seasons (~15%). The average hourly N_{acc} concentrations ranged between 4200 cm^{-3} (monsoon midday) to 26200 cm^{-3} (winter evening). For all seasons, the hourly averaged hourly N_{acc} concentrations had a morning (07:00) peak and an evening (20:00–22:00) peak, with lowest concentrations during the daytime (13:00–15:00). In addition to emitting in the Aitken mode, biomass burning and older diesel vehicles emit in the accumulation mode as well ~~(Chen et al., 2017; Morawska et al., 1998)~~(Chen et al., 2017; Paasonen et al., 2016; Kumar et al., 2010). Additionally, particles from fresh vehicular emissions can grow rapidly from the Aitken mode at the tailpipe to accumulation mode at roadside and ambient locations (Ning et al., 2013). We speculate that the traffic-related particles in Delhi may get smaller as India's vehicle fleet (especially heavy-duty trucks) is upgraded (Guttikunda and Mohan, 2014; Zhou et al., 2020a).

Overall, UFP contributed to ~65% of the PN concentrations for winter and autumn. The UFP fraction of PN concentrations was relatively higher for spring (75%), summer (78%), and monsoon (80%). The average hourly UFP concentrations ranged between 17200 cm^{-3} (autumn midday) to 52500 cm^{-3} (winter evening). For all seasons, UFP concentrations had peaks in the morning (07:00–09:00) and the evening (19:00–21:00). For the relatively warmer seasons (summer, monsoon, and spring), there were additional peaks during the daytime (11:00–13:00). The UFP fraction of PN (UFP/PN) was generally the highest during the daytime in spring, summer, and monsoon (~85%). Conversely, some of the lowest UFP/PN levels were observed during the nighttime of winter and autumn (~55%). The UFP/PN levels observed in Delhi are generally lower than those in cleaner cities (Hussein et al., 2004; Rodríguez et al., 2007; Putaud et al., 2010) and even lower compared to some other polluted ones where mass concentrations often exceeded ~100 μg m^{-3} (Whitby et al., 1975; Laakso et al., 2006; Wu et al., 2008). As a

result of the relatively large fraction of PN being constituted of non-UFP (N_{acc}) in Delhi, the median diameter size observed were often much larger than those observed in these cleaner cities.

The hourly averaged median diameters were largest during nighttime of winter and autumn, reaching up to ~90 nm. The smallest hourly averaged median diameters (~40 nm) were observed during the daytime of spring, summer, and monsoon. As with the concentrations and fractions of the PN modes, the median diameters also had sharp seasonal and diurnal variations. For winter and autumn, the hourly averaged median diameters ranged between 50 nm during the late afternoon (16:00) to 90 nm during the late night/early morning (03:00). The median diameters were the smallest for the relatively less polluted warmer months (summer, monsoon, and spring), with the hourly averaged median diameters ranging between 35 nm (12:00) to 75 nm (03:00). For comparison, the median diameter in Helsinki (calculated for ~12–560 nm) was ~30 nm (Hussein et al., 2004). In Los Angeles, the median diameters up to 150 m from a freeway (our site is ~150 m from an arterial road) were <50 nm (Zhu et al., 2002). We found that for all seasons, the nighttime had larger particles than the daytime. For winter and autumn, the average median diameters were ~65 nm during the day and ~80 nm during the night. For the spring, summer, and monsoon, the average median diameters were ~40 nm during the day and ~65 nm during the night.

In Fig. 6, we present the average observed PSD evolving over the day for each season as a heatmap. For the spring, summer, and monsoon the average seasonal heatmap indicates daytime new particle formation. However, winter and autumn had lower concentrations of the smaller particles that are generally associated with new particle formation. In the following section (Sec. 3.3) we explore the role of coagulation scavenging during polluted periods in selectively suppressing concentrations of smaller particles and resulting in an increase in the median diameter size.

3.3 Coagulation scavenging

The evolution of ambient particle size distributions reflects the complex interplay between emissions, atmospheric dilution, and a wide range of aerosol dynamic processes including new particle formation, evaporation/condensation, and coagulation. In this section we explore the role of coagulation scavenging in suppressing both the existing UFP concentrations and new particle formation during periods with high aerosol mass loadings corresponding to elevated N_{acc} levels. During the polluted winter and autumn seasons, the daytime UFP levels were often lower than those during the same period in the warmer months.

In Fig. 7 we present PSD heatmaps for an extremely polluted (high aerosol mass loading) period (Feb 1–5, 2017). During the polluted episode (Fig. 7 (a)), the $PM_{0.56}$ concentrations ranged between 100 –400 and 400 μg m^{-3} and the PN concentrations between 25000 –80000 and 80000 cm^{-3}. The peak PN levels were observed during the evening traffic rush hours. Even though PN concentrations decreased during the late evening (~8–11 PM), the $PM_{0.56}$ levels kept continuously increasing all night. The N_{nuc} levels decreased during these polluted evening periods even as N_{ait} and N_{acc} levels increased (Fig. S13). This dynamic suggests that coagulation may be acting as a strong control on concentrations of smaller particles, even as emissions are high. The characteristic timescale for particles smaller than 15 nm to be lost to coagulation scavenging by larger particles was less than 15 minutes for these polluted evenings. The size of the particles rapidly increased from the evening to the nighttime (growth rates up to ~10 nm h^{-1}) and coagulation may be one of the mechanisms that contributes to this rapid growth.

Since coagulation only affects particle number concentrations, and not mass concentrations, $PM_{0.56}$ increased into the night, possibly owing to a combination of increased sources (e.g., nocturnal truck traffic, biomass burning for cooking and heat) and decreasing ventilation ~~(Guttikunda and Gurjar, 2012; Guttikunda and Calori, 2013; Bhandari et al., 2020a)~~(Guttikunda and Gurjar, 2012; G The PN concentrations dropped after the evening traffic rush hours and only increased again during the morning traffic rush hours.

5   We followed Westerdahl et al. (2009) to calculate the coagulation timescales corresponding to the observed PSD (Sec. 2.3). When a small particle coagulates onto a larger particle, the small particle is lost and the big particle scarcely grows. In Fig. 8 we have presented the diurnal and seasonal profiles for the modeled coagulation timescales for a 15, 30, and 100 nm particle. For the nucleation mode particle (15 nm), the timescales ranged from a few minutes for the polluted periods (mornings and evenings of cooler seasons) to ~1 h for the relatively less polluted periods (midday of warmer seasons). The corresponding

10   range for an Aitken mode particle (30 nm) was ~~from~~ ~1–4 h. Finally, for an accumulation mode particle (100 nm), coagulation was not a significant sink owing to the extremely long timescales (tens of hours). Advection timescales across Delhi are ~3–5 h based on a length scale of ~40 km at typical wind speeds.

Coagulation scavenging of UFP on to the larger accumulation mode particles could explain the suppression in UFP concentrations for polluted periods which have high accumulation mode concentrations. The high aerosol surface area from particles

15   in the accumulation mode, in addition to contributing to most of the mass, can act as a coagulation sink for the smaller particles. The scavenging of UFP in Delhi during the polluted periods could explain the higher median particle diameters than those usually observed in other urban environments (Salma et al., 2011). In Table 2, we compare characteristic condensation sink (~~SO₂~~H_2SO_4) and coagulation sink (for 1, 5, 10, 15, 30, and 100 nm particles) for Delhi (least and most polluted) with other cities (clean and polluted). Based on the coagulation timescales for these polluted periods, for a 1 h period coagulation

20   scavenging could result in removal of ~85% of the 10 nm particles, ~50% of the 30 nm particles, and ~10% of the 100 nm particles present at the beginning of the hour. The coagulation sink for UFP in Delhi during the polluted periods was ~20 times larger as compared to a clean city (Helsinki). Even for least polluted periods in Delhi, the coagulation sink for UFP was ~4 times larger than Helsinki.

3.4   New particle formation

25   For the less polluted seasons (Fig. 6), we observed a sharp increase in the concentration of nucleation mode particles during middays suggesting new particle formation. In Fig. 7 (b) we present PSD heatmaps for a relatively less polluted period (Apr 7–11, 2017). During some days for this period we observed new particle formation and growth. The growth of particles in the nucleation mode was especially prominent for the latter two days of this episode when the $PM_{0.56}$ concentrations were almost half the $PM_{0.56}$ concentrations of the first two days and consequently both the condensation and coagulation sinks (across

30   particle sizes) also decreased by ~50%. We observed "banana-shaped" new particle formation and growth events during these relatively clean conditions which were consistent with growth of atmospheric nanoparticles observed elsewhere (Kulmala and Kerminen, 2008). The growth ~~rates~~ rate of the nucleation mode particles ~~for these growth events were ~4~~ based on the average

PSD for these clean conditions was ~5 nm h^{-1}. It should be noted that our SMPS measurements had a lower size ~~cut~~ cut-off of 12 nm, implying that the particles in the nucleation mode had already grown before being detected.

Growth rates depend on particle size, concentrations of condensable gases (H$_2$SO$_4$, highly oxygenated molecules, etc.), and condensation and coagulation sinks, and they can range from 0.1 to 10s of nm h^{-1} across various environments (Kerminen et al., 2018; Niem We did not observe "banana-shaped ~~growth patterns very often~~" new particle formation and growth events during the polluted winter or autumn seasons as the growth phase was probably disrupted by coagulation scavenging. For example, ~~even for the least polluted monsoon~~ during winter, coagulation timescales for a 15 nm particle were ~~~1~~ <1 h (Fig. 8). ~~In contrast, the new particle formation related growth rate observed are generally between 1–10 nm h^{-1} (Bianchi et al., 2016; Kulmala et al., 2004), with growth~~ Growth rates for polluted megacities also usually slower than ~~5~~ 10 nm h^{-1} ~~(Zhao et al., 2018, and references therein). So, as has been shown in studies from other polluted environments,~~ (Zhou et al., 2020b; Chu et al., 2019). It is likely that a nucleation mode particle is more likely to coagulate ~~on to larger particles~~ with a larger particle than to grow ~~up~~ to the Aitken/accumulation mode. Overall, the aerosol dynamics are a complex interplay of both new particle formation and coagulation scavenging, with nucleation mode particles in highly polluted environments generally susceptible to ~~getting 'lost' onto~~ scavenging by the accumulation mode particles.

Large accumulation mode concentrations also act as a strong condensation sink (Table 2), causing vapors to condense on to existing particles instead of forming new particles (Mönkkönen et al., 2004a). During the most polluted time in Delhi (generally 8–9 PM during the winter), the condensation sink was 20 times larger than Helsinki (clean city) (Hussein et al., 2004). Even for the least polluted period in the Delhi (generally 2–3 PM during the monsoon), the condensation sink was ~4 times larger than Helsinki. The condensation sink for the polluted periods was somewhat higher than Beijing (polluted city) (Laakso et al., 2006), but within the same order of magnitude. For spring, summer, and monsoon, we estimated the condensation sink to be more than an order of magnitude lower than the winter and autumn. Consequently, unlike winter and autumn, we observed an increase in daytime UFP concentrations (around 12–30 nm) for the relatively less polluted months which was consistent with daytime new particle formation from nucleation events (Kulmala et al., 2004; Brines et al., 2015). Based on the survival parameter (P = (CS/10^4 s^{-1})/(GR/nm h^{-1})) defined in Kulmala et al. (2017), new particle formation would not be expected even during some of the least polluted periods in Delhi. Why we observe any new particle formation events in Delhi—similar to observations from megacities in China—is still not well understood (Kulmala et al., 2017).

3.5 Role of meteorology

In Gani et al. (2019) we showed that meteorology—specifically change in ventilation—was an important factor in driving the seasonal aerosol mass variations observed in Delhi. However, PN levels did not show similar seasonal variation (Fig. 2). In Fig. S11, we have presented the 2-week averages for ventilation and PN concentrations (by mode and total). N$_{acc}$ concentrations decrease as the ventilation coefficient increases. Since accumulation mode particles constitute most of the submicron mass, decrease in N$_{acc}$ concentrations with increasing ventilation is consistent with our previous analysis that showed decrease in mass loadings with increasing ventilation. N$_{ait}$ concentrations seem to have a smaller slope than accumulation mode concentrations. N$_{nuc}$ concentrations increased with increasing ventilation, consistent with new particle formation and/or less

coagulation scavenging. Comparing rainy and non-rainy days during the monsoon, our observations suggest that rain reduces the accumulation mode concentrations (Fig. S12) as has been observed in other studies (Luan et al., 2019; Hussein et al., 2018). However, we do not observe rain to cause a clear reduction in either nucleation or Aitken mode concentrations. Parametrization studies suggest that rain is more effective in scavenging nucleation mode particles as compared to Aitken mode particles (Pryor et al., 2016; Laakso et al., 2003). Lower accumulation mode concentrations following rain events imply lower condensation and coagulation sinks. While we cannot fully reconcile our observations here with theory, possible explanations include (i) differential water solubility by particle size and age, or (ii) that the rain washout of the nucleation mode particles is 'balanced' by the reduction in the condensation and coagulation sink due to rain washout of accumulation mode particles.

The lower sensitivity of PN to ventilation changes can be explained by the dissimilar sources and atmospheric process that contribute to PN and PM levels. While PN is mostly comprised of UFP, accumulation mode particles constitute most of the fine aerosol mass. Traffic, cooking, and nucleation events contribute to urban UFP. A large fraction of accumulation mode particles in contrast result from biomass burning and aging of aerosol which result in particles often larger than 100 nm (Janhäll et al., 2010). Periods with lower ventilation that result in high aerosol mass loadings also create a large coagulation sink for UFP (Sec. 3.3). While coagulation suppresses the PN levels, the aerosol mass remains conserved. While a decrease in ventilation should initially increase concentrations for all particle sizes similarly, coagulation causes the smaller UFP to be selectively 'lost'—causing a larger increase for aerosol mass compared to number. Conversely, periods with high ventilation (daytime of warmer months) also have more nucleation events resulting in increased UFP concentrations, but with almost no contribution to aerosol mass (Brines et al., 2015). In addition to large condensation sinks from existing particles (Sec. 3.4), it is possible that the elevated RH during the cooler winter and autumn (Fig. 1) could further suppress atmospheric nucleation. It has been observed in both clean and polluted environments that the RH during new particle formation event days tends to be lower compared to non-event days (Kerminen et al., 2018; Dada et al., 2017; Hamed et al., 2011). Overall, ventilation strongly influences aerosol mass loadings in Delhi, with higher ventilation often resulting in reduction of PM concentrations and vice versa. However, we do not observe such a simple relationship between ventilation and PN levels. The complex interplay of sources, aerosol dynamics, and atmospheric processes as discussed above make PN levels less sensitive to meteorological changes as compared to aerosol mass loadings.

3.6 Lessons from multi-modal PSD fitting

In this section we present the parameters obtained by fitting multi-modal lognormal distributions on the seasonal and characteristic time-of-day averages of the observed PSD (Table 3). For most seasons and times of day, the PSD were bimodal. Even when there was a third mode, it contained less than 10% of the total PN concentration. While the smaller mode (ultrafine mode) ranged from 20 to 40 nm depending on season and time of day, the larger mode (accumulation mode) ranged from 80 to 120 nm. For all seasons the late night ultrafine mode concentrations contributed to less than half of the total

PN concentration. This observation can be explained by fewer sources of UFP—less traffic and no nucleation events—and a large coagulation sink from nighttime aerosol mass loadings (from accumulation mode) which are often high in part due to unfavorable nighttime meteorology (Gani et al., 2019). Delhi traffic usually peaks around 8–11am in the morning and 5–9pm in the evening. Restrictions on daytime heavy duty truck traffic have resulted in increased nocturnal truck traffic after 10pm (Mishra et al., 2019; Guttikunda and Calori, 2013). The accumulation mode during late nights had median diameters between 100 nm for the relatively less polluted summer and monsoon to 120 nm for spring, winter, and autumn.

During the morning rush hour, the median diameter of the ultrafine and accumulation mode were the smallest for the summer (21, 100 nm for ultrafine and accumulation mode) and monsoon (23, 110 nm) compared to spring (28, 122 nm), autumn (35, 120 nm), and winter (39, 122 nm). Autumn had three modes (35, 120, 234 nm) with the ultrafine mode contributing to 58%, accumulation mode to 34% and the third mode contributing to only 8% of the total PN concentration. For winter during the same period, half of the PN concentrations were in the ultrafine mode (~40 nm) and the other half in the accumulation mode (~120 nm). The fraction of PN in ultrafine mode during the same period was relatively higher for the other seasons which had less aerosol mass loadings compared to winter, with the least polluted monsoon having 66% of the PN in the ultrafine mode.

The midday PSD were bimodal for winter (26, 125 nm), autumn (29, 122 nm), spring (19, 100 nm), and summer (26, 111 nm). For monsoon the PSD was trimodal (27, 90, 177 nm), with only 6% of the PN fraction in the third mode. Generally, the midday hour had some of the highest fraction of PN concentrations in the ultrafine mode, especially for summer (80%), monsoon (83%), and spring (81%). The corresponding levels while much lower for the more polluted autumn (56%) and winter (53%), were higher than most other times of day within those seasons. Even in absolute terms, summer, monsoon, and spring had some of the highest PN concentrations in the ultrafine modes. The high ultrafine mode concentrations—both magnitude and PN fraction—is potentially from new particle formation (Kulmala et al., 2004). We observe characteristic new particle formation followed by some growth for the warmer seasons (Fig. 6). However, during the more polluted winter and autumn, daytime new particle formation is not observed—potentially because of strong coagulation and condensation sink (Sec 3.4).

During the evening rush hour, the PSD were bimodal for winter (35, 88 nm), autumn (39, 103 nm), spring (28, 80 nm), and summer (30, 89 nm). For monsoon the PSD was trimodal (27, 76, 203 nm), with only 5% of the PN fraction in the third mode. The relatively smaller median diameter for the accumulation mode during the traffic rush hour is consistent with particles from fresh vehicle exhaust—both gasoline and diesel—being smaller than those from biomass burning and other aged aerosol. While gasoline engines emit in the ~20–60 nm range, ~~diesel engines emit in the ~20–130 nm size range (Ristovski et al., 1998; Morawska et al., 1998)~~ some of the older diesel engines still prevalent in India can emit larger accumulation mode particles (Kumar et al., 2010). Particles from biomass burning are in the ~50–200 nm size range (Chen et al., 2017; Janhäll et al., 2010). For the warmer periods which have relatively less biomass burning emissions, the ultrafine modes contribute to relatively higher fraction of PN—especially during traffic rush hours. These findings are consistent with studies from cities in high-income countries where traffic and nucleation events are considered the major source of UFP (Brines et al., 2015).

4   Conclusions

We used continuous, highly time-resolved and long-term data to provide a detailed seasonal and diurnal characterization of Delhi's PN concentrations. The number concentrations for each mode (nucleation, Aitken, and accumulation) varied dynamically by season and by time of day. While Delhi experiences PM levels 10 times higher than cities in North America and Western Europe, PN levels were comparable to those observed in urban sites in relatively cleaner (in terms of aerosol mass) cities~~where PN concentrations have generally observed to be around 10000–60000 cm^{-3} (Kumar et al., 2014)~~. Observations from other polluted cities (e.g., in China) also show that high aerosol mass does not necessarily imply similarly high PN concentrations ~~(Laakso et al., 2006; Wu et al., 2008; Shen et al., 2011)~~(Shen et al., 2011; Wu et al., 2008; Laakso et al., 2006). Furthermore, the seasonal variability of PN was much less than that of the PM measured at our site. While it is generally assumed that UFP constitute most of the PN concentrations, we observed that large number of accumulation particles—that constitute most of the fine aerosol mass—contributed to almost half of the PN concentrations for some of the extremely polluted periods. UFP concentrations were found to be lower during periods with some of the highest mass concentrations.

We show that the lack of proportionality between aerosol mass and number concentrations result from rapid coagulation of UFP, especially during periods with high accumulation mode concentrations. Furthermore, the accumulation mode particles can also act as a strong condensation sink, causing vapors to condense onto existing particles instead of forming new particles. Even though coagulation does not affect mass concentrations, it can significantly govern PN levels with important health and policy implications. Furthermore, implications of a strong accumulation mode coagulation sink for future air quality control efforts in Delhi are that a reduction in mass concentration, may not produce proportional reduction in PN concentrations. Long term continuous observations of PSD from Delhi will be able to provide important insights into the role of sources and atmospheric processes that drive aerosol number concentrations.

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Table 1. Day and night summary of observations derived from the PSD and meteorological parameters. Arithmetic mean used for all species and parameters, except wind direction for which we used median to estimate its central tendency.

	Winter		Spring		Summer		Monsoon		Autumn	
	Day	Night	Day	Night	Day	Night	Day	Night	Day	Night
N_{nuc}[a] ($\times 10^3$ cm^{-3})	7.6	5.0	15.8	9.3	13.4	7.8	13.7	6.9	6.1	3.8
N_{ait}[a] ($\times 10^3$ cm^{-3})	21.3	31.4	19.7	27.1	19.6	24.2	18.0	17.6	17.1	20.9
N_{acc}[a] ($\times 10^3$ cm^{-3})	13.4	22.8	8.4	14.8	7.0	12.3	4.9	7.7	9.8	15.8
UFP[b] ($\times 10^3$ cm^{-3})	28.9	36.4	35.5	36.4	33.0	32.0	31.7	24.5	23.2	24.7
PN[c] ($\times 10^3$ cm^{-3})	42.3	59.2	43.9	51.2	40.0	44.3	36.6	32.2	33.0	40.5
UFP/PN	0.68	0.62	0.81	0.71	0.83	0.72	0.87	0.76	0.70	0.61
Median D_p (nm)	63.1	81.2	38.5	64.1	43.2	63.4	39.5	58.6	63.1	80.3
SA ($\times 10^3$ μm^2cm^{-3})	2.04	3.24	1.25	2.12	0.99	1.65	0.74	1.06	1.51	2.21
Vol (μm^3cm^{-3})	84.1	128	47.5	81.0	34.3	58.6	25.9	37.3	62.1	87.1
$PM_{0.56}$[d] (μg ^{3}m^{-3})	135	204	76.0	130	54.9	93.7	41.4	59.8	99.4	139
Temperature (°C)	17	13	26	21	35	31	32	29	28	23
Relative Humidity (%)	60	78	42	59	34	43	71	81	49	67
Wind Speed (ms^{-1})	2.7	2.6	3.2	2.6	3.8	2.9	3.4	2.5	2.5	2.1
Wind Direction (°N)	300	300	300	300	270	270	250	190	300	250
PBLH (m)	920	340	1800	1000	2400	1600	1600	460	1460	880

[a]The modes are based on SMPS observations—nucleation (12 < D_p < 25 nm), Aitken (25 < D_p < 100 nm) and accumulation (100 < D_p < 560 nm) modes. [b]UFP = N_{nuc} + N_{ait}. [c]PN = UFP + N_{acc}. [d]Estimation of $PM_{0.56}$ concentrations were based on the volume concentrations observed by the SMPS and assuming particle density to be 1.6 g cm^{-3}.

Table 2. Characteristic coagulation and condensation sinks for Delhi and comparison with those calculated ~~for~~ using PSD in literature from other cities.

		DAS (our study)		Literature		
		Most polluted[a]	Least polluted[b]	Delhi[c]	Beijing[dc]	Helsinki[ed]
Condensation sink (s^{-1})	~~SO$_2$~~H$_2$SO$_4$	~~2.0~~1.7×10^{-1}	~~4.1~~3.5×10^{-2}	~~1.7~~1.4×10^{-1}	~~1.3~~1.1×10^{-1}	~~9.7~~8.2×10^{-3}
Coagulation sink (s^{-1})	1 nm	8.7×10^{-2}	1.8×10^{-2}	7.3×10^{-2}	5.7×10^{-2}	4.3×10^{-3}
	5 nm	5.7×10^{-3}	1.3×10^{-3}	4.6×10^{-3}	3.4×10^{-3}	3.1×10^{-4}
	10 nm	1.8×10^{-3}	4.2×10^{-4}	1.4×10^{-3}	1.0×10^{-3}	1.1×10^{-4}
	15 nm	9.2×10^{-4}	2.3×10^{-4}	7.2×10^{-4}	5.2×10^{-4}	5.6×10^{-5}
	30 nm	3.2×10^{-4}	7.4×10^{-5}	2.4×10^{-4}	1.7×10^{-4}	1.9×10^{-5}
	100 nm	3.7×10^{-5}	6.4×10^{-6}	3.0×10^{-5}	7.2×10^{-5}	1.5×10^{-6}

[a] Average PSD for Winter 8–9 PM. [b] Average PSD for Monsoon 2–3 PM. [c] PSD from Fig. 2 in Laakso et al. (2006). [d] PSD from Table 1 in Hussein et al. (2004).

Table 3. Multi-lognormal fitting parameters for the average particle size distribution of each season and characteristic times of day.

|		Mode 1				Mode 2				Mode 3			
		D_p (nm)	σ_g	N_{tot} (×10³ cm⁻³)	%PN	D_p (nm)	σ_g	N_{tot} (×10³ cm⁻³)	%PN	D_p (nm)	σ_g	N_{tot} (×10³ cm⁻³)	%PN
2–3 AM	Winter	35	1.8	15.8	31	122	1.8	34.5	69	—	—	—	—
	Spring	28	1.8	20.0	43	122	1.8	26.2	57	—	—	—	—
	Summer	23	1.6	9.1	26	100	1.8	26.0	74	—	—	—	—
	Monsoon	26	1.8	10.2	38	101	1.8	16.7	62	—	—	—	—
	Autumn	35	1.8	10.4	28	122	1.8	26.3	72	—	—	—	—
8–9 AM	Winter	39	1.9	31.7	50	122	1.9	31.8	50	—	—	—	—
	Spring	28	1.7	37.2	61	122	1.8	24.1	39	—	—	—	—
	Summer	21	1.6	25.4	56	100	1.9	20.0	44	—	—	—	—
	Monsoon	23	1.6	23.5	66	110	1.8	12.2	34	—	—	—	—
	Autumn	35	1.7	26.4	58	120	1.6	15.2	34	234	1.6	3.6	8
2–3 PM	Winter	26	1.7	17.1	53	125	1.8	15.0	47	—	—	—	—
	Spring	19	1.8	41.9	81	100	2.0	9.7	19	—	—	—	—
	Summer	26	1.9	35.5	80	111	1.7	8.7	20	—	—	—	—
	Monsoon	27	1.7	36.5	83	90	1.4	4.7	11	177	1.6	2.7	6
	Autumn	29	1.7	14.3	56	122	1.9	11.2	44	—	—	—	—
8–9 PM	Winter	35	1.7	20.8	25	88	1.9	63.0	75	—	—	—	—
	Spring	28	1.9	38.3	55	80	2.0	31.8	45	—	—	—	—
	Summer	30	1.8	35.9	57	89	1.9	27.2	43	—	—	—	—
	Monsoon	27	1.6	25.4	56	76	1.7	17.7	39	203	1.6	2.2	5
	Autumn	39	1.9	24.8	48	103	1.9	26.8	52	—	—	—	—

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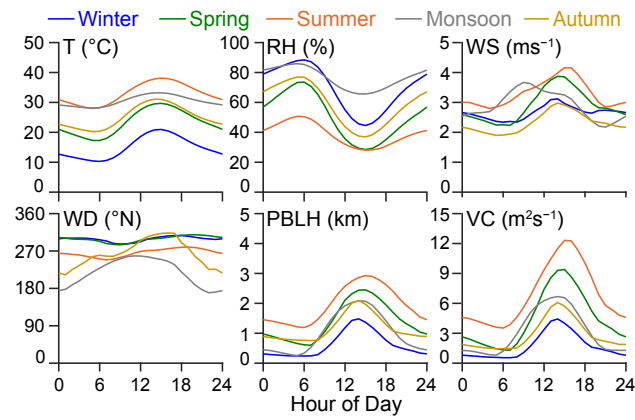
Figure 1. Diurnal profiles of meteorological parameters (temperature, relative humidity, wind speed, wind direction, PBLH and VC) by season. Average values by season and hour of day are presented for all parameters except wind direction. The median value is presented for wind direction. Ventilation coefficient (VC) = PBLH × wind speed.
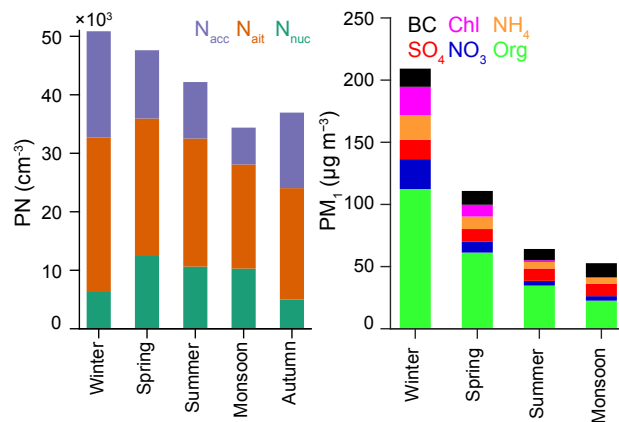
Figure 2. Average PN levels for each season by mode. The modes are based on SMPS observations—nucleation ($12 < D_p < 25$ nm), Aitken ($25 < D_p < 100$ nm) and accumulation ($100 < D_p < 560$ nm) modes. The PM$_1$ plot is reproduced from Gani et al. (2019) to illustrate seasonal variation in aerosol mass loadings. The PM$_1$ species are organics (Org), chloride (Chl), ammonium (NH$_4$), nitrate (NO$_3$), sulfate (SO$_4$), and black carbon (BC). While we had SMPS data for all seasons, we did not collect PM composition data during autumn due to instrumentation (ACSM) downtime.
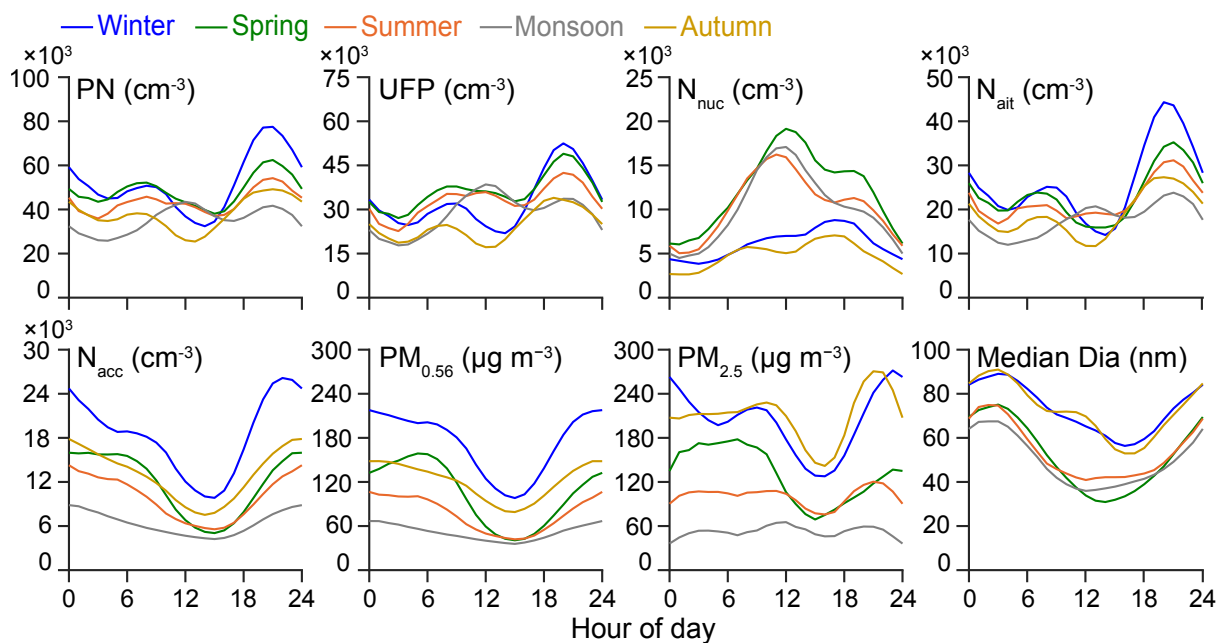
Figure 3. Average diurnal and seasonal variations for PN, UFP ($D_p < 100$ nm), N_{nuc} ($D_p < 25$ nm), N_{ait} ($25 < D_p < 100$ nm), N_{acc} ($D_p > 100$ nm), and median particle diameter. These averages are based on the observed SMPS data ($12 < D_p < 560$ nm). Also included are the average diurnal and seasonal variations of the mass concentrations estimated from the observed SMPS data ($PM_{0.56}$) and the $PM_{2.5}$ concentrations from a regulatory monitor (DPCC, R.K. Puram, 3 km from our site).
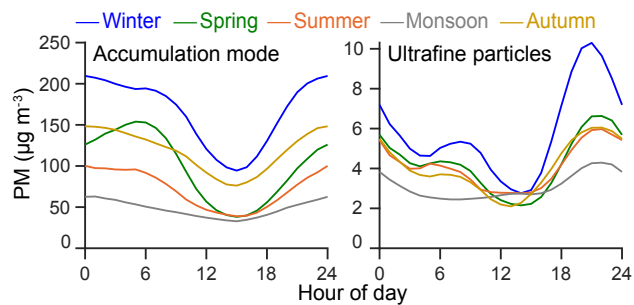
Figure 4. Average mass concentrations of observed accumulation mode (100–560 nm) and ultrafine particles (<100 nm) by season and time of day. Estimation of mass concentrations based on assumed particle density of 1.6 g cm^{-3}.
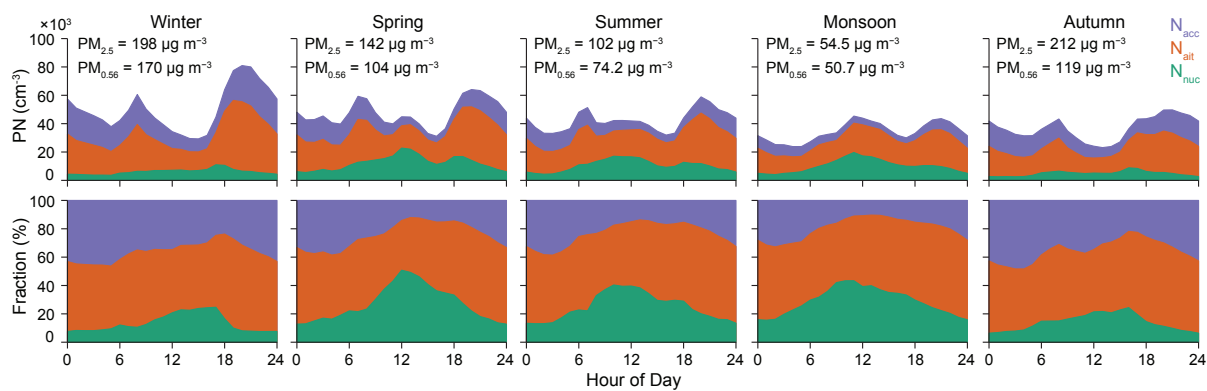
Figure 5. Stacked average absolute and fractional diurnal profiles of N_{nuc} ($D_p < 25$ nm), N_{ait} ($25 < D_p < 100$ nm), and N_{acc} ($D_p > 100$ nm) by season. These averages are based on the observed SMPS data ($12 < D_p < 560$ nm). We also include seasonal average concentrations for the PM$_{2.5}$ concentrations from a regulatory monitor (DPCC, R.K. Puram, 3 km from our site) and mass concentrations estimated from the observed SMPS data (PM$_{0.56}$).
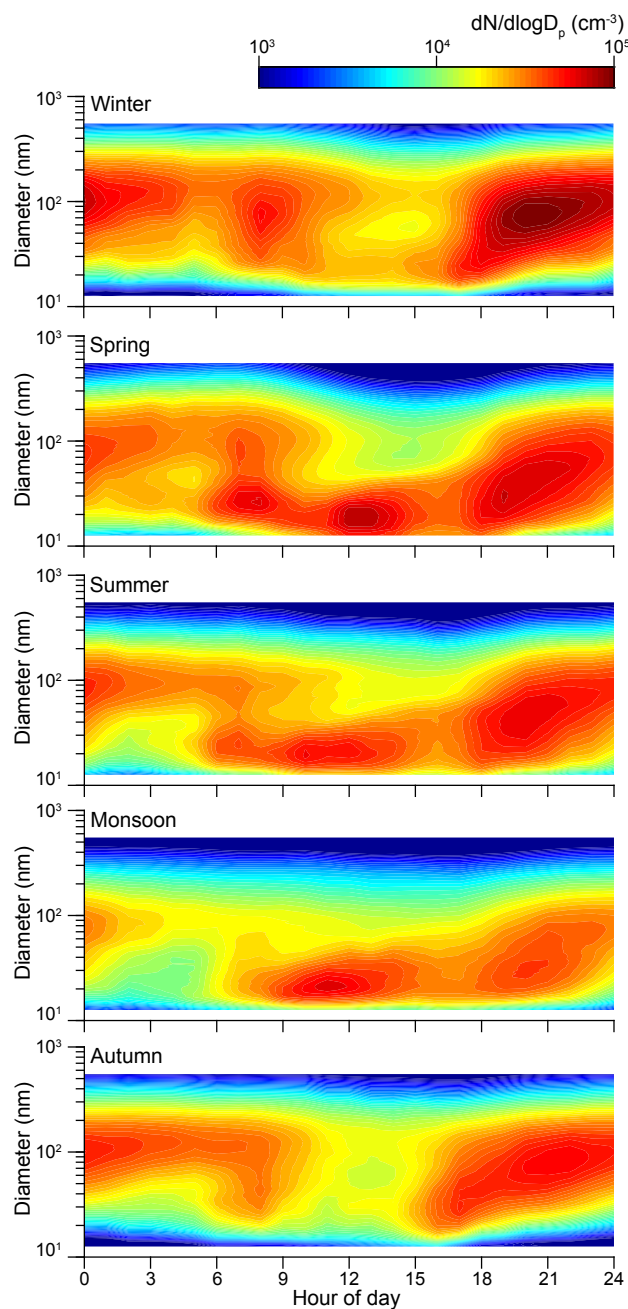
Figure 6. Heatmap for particles between 12 ~~560~~ and 560 nm averaged for each season.
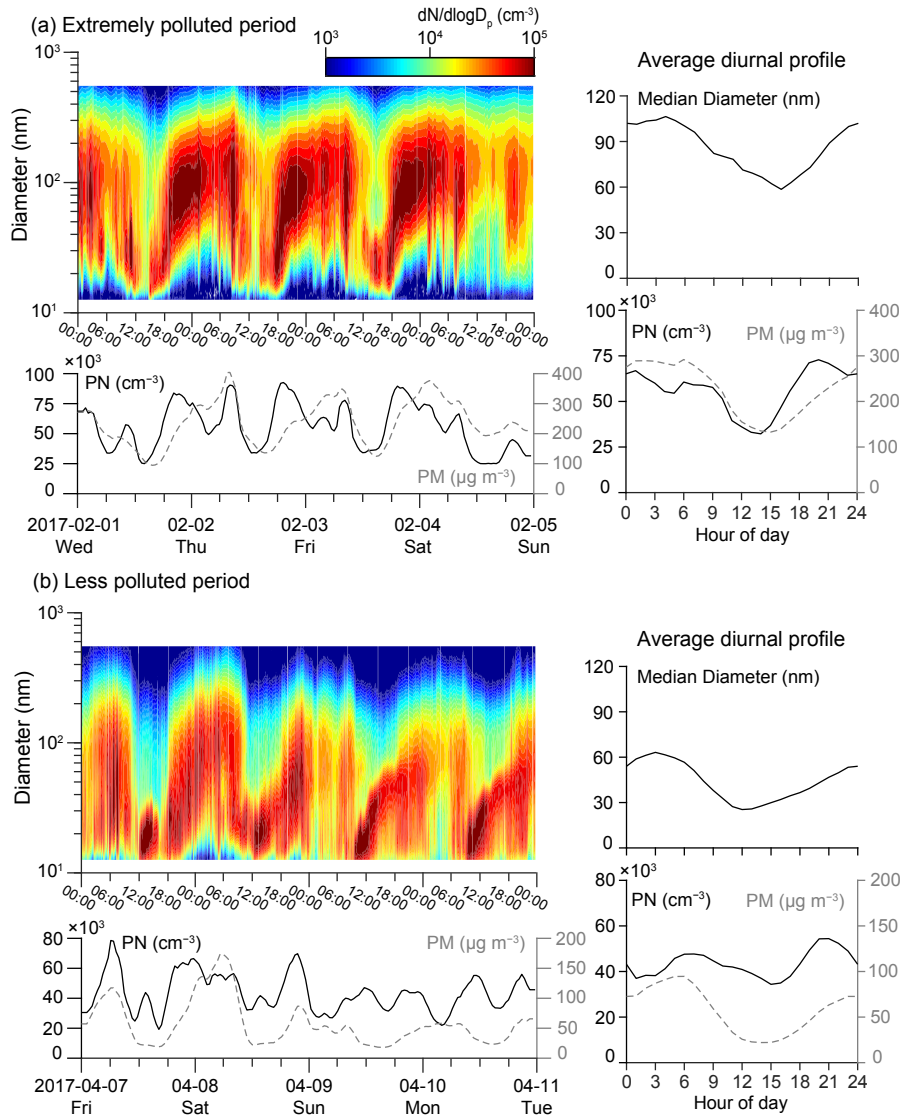
Figure 7. Heatmap showing the evolution of the PSD for a (a) polluted period with prominent coagulation scavenging and (b) less polluted period with some new particle formation and growth. Bulk PN and PM concentrations for the same period as the heatmap are also presented for both periods. The average diurnal profile of the median diameter along with the PN and PM concentrations over the two periods are presented in the right panels. Estimation of PM concentrations were based on the volume concentrations observed by the SMPS and assuming particle density to be 1.6 g cm^{-3}.
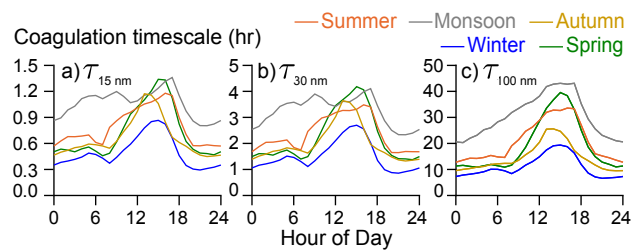
Figure 8. Coagulation timescale (τ) for 15, 30 and 100 nm particles by season.