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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Particle number concentrations and size distribution in a polluted megacity: The Delhi Aerosol Supersite study" *by* Shahzad Gani et al.

Anonymous Referee #2

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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.

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.

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).

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.

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).

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).

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).

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

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)?

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.

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