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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Simultaneous measurements of new particle formation in 1-second time resolution at a street site and a rooftop site" *by* Yujiao Zhu et al.

Anonymous Referee #5

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Fast-response measurements of particle number size distributions of aerosol $\geq$8 nm diameter have been made at a street canyon and nearby rooftop site. The authors selectively report specific days of data from a small dataset, and draw many tentative conclusions concerning mechanisms of new particle formation (NPF) which are difficult to justify given the small dataset and the extent to which it is over-interpreted. The introduction quite reasonably states that "it is critical to evaluate the effects of nucleating species other than sulfuric acid and the dependence of NPF on pre-existing particles in the atmosphere". This is an excellent objective but unfortunately the paper does nothing to answer the question about other nucleating species, and does not event provide clear answers concerning the role of sulfuric acid.

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One of the key elements towards interpretation of this dataset in relation to nucleation and growth is the role of sulfuric acid, which ideally would have been measured. However, as measurements were not available, an old parameterisation is used to estimate H2SO4 vapour concentrations in which the H2SO4 formation rate is described by the product of SO2 concentration and global solar radiation. This may be adequate for situations in the background troposphere where ozone photolysis is the predominant source of hydroxyl radical, but many studies have now shown that in polluted atmospheres such as Beijing, other processes such as photolysis of HONO and HCHO, and ozone-alkene reactions are far more important sources of hydroxyl, and equation 4 is unlikely to be a reliable means of calculation of [H2SO4].

The differences in behaviour between the sites are interesting, and if correctly interpreted could give useful insights into NPF in polluted atmospheres. However no measurements were made of potentially condensing species, or their precursors other than SO2, and the latter was measured at only one site with the unproven assumption that concentrations of SO2 were the same at both sites. Much is made of the rates of change of particle number concentrations, but the effects of wind direction changes upon concentrations in the street canyon (which can be large) do not appear to have been considered. The methods used for subtraction of fresh traffic emissions are highly questionable, and no use is made of gaseous pollutant data (e.g. NOx) which would be a strong covariate of PNC from road vehicles.

The points above justify a major reappraisal of the data, and the development of far less ambitious conclusions. Other points which need to be addressed include:

(a) The introduction lists a number of organic acids as examples of vehicle-emitted organic compounds. Most of these have far more major secondary sources, or are present in cooking emissions, with little if any arising from road traffic.

(b) Some ill-informed statements are made about the (currently uncertain) effects of exposure to ultrafine particles. These particles do not lead to “destruction of the res-

piratory system" and the statement that "newly formed particles inside a street canyon may become toxic when vehicle-release organics is involved in the nucleation process" is not supported by references.

(c) There is no information on quality assurance beyond an intercomparison between the two FMPS, and no consideration of how size-dependent particle losses in the inlet system affect measured size distributions.

(d) Equation (3) differs from that in the nucleation protocol paper of Kulmala et al. (2012) by a factor of two, which needs to be explained.

(e) A clear definition is needed for the "maximum increase of nucleation mode PNC (NMIoNP)" which is much used in the data analyses.

(f) The authors should establish that their Class II particles arise from an NPF event, rather than an emission source.

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