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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.

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Fast-response measurements of particle number size distributions of aerosol ≥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.

Response: The authors didn't selectively present the data. All simultaneous measurements on particle number concentrations during the springtime and wintertime campaigns have been included in this study. At the street site, the reduced NPF events always occurred in the springtime and the enhanced NPF events always occurred in the wintertime. This will be highlighted in revision.

The authors would like to believe the sufficiency and uniqueness of evidences are critical to evaluate the quality of scientific studies. This is because the number of cases either for gravitational wave observation in 2016 or a recent NPF study reported by Bianchi et al. (2016) was even smaller than those presented in this study.

This study is definitely not a first study for NPF events and all analyses build on previously well-established knowledge, particularly the progressing in the last few years. The authors did provide unique evidences to analyze the effects of nucleating species other than sulfuric acid at street site. The authors also agree that more analyses should be added so that they can be easily understood. For example, in revision, the authors will add "Considered 1) formation rate of new particles, e.g., J=kNucOrg[H2SO4]m[NucOrg]n (Zhang et al., 2012), 2) the subsequent particle growth, and 3) H2SO4 vapor to be necessary for nucleation in ambient air except at sea beach, two scenarios are considered. One is: H2SO4 vapor is relatively sufficient against NucOrg and J8 is thereby mainly determined by availableness of NucOrg. A good correlation is theoretically expected for J8 and NMIoNP. The other scenario is: NucOrg is relatively sufficient against H2SO4 vapor and J8 is thereby mainly determined by availableness of H2SO4 vapor. J8 could be high, but the total yield of new particles could be low because of a rapid consumption of H2SO4 vapor. A poor or no correlation is theoretically expected for J8 and NMIoNP."

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. How-

ever, 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].

Response: The authors would like to believe that the reviewer may capture a piece of information about air pollution in Beijing, but it is hard to say the piece reflecting the full picture.

During the periods of NPF events in this study, the air mass at the sampling site was less polluted or even clear. The NPF events occurred under the north or northwest wind direction with wind speed >4m/s. The north and northwest directions of the sampling site subject to mountain areas have a high percentage of land-covered forests. The north or northwest wind carried less polluted or even clear ambient air to the sampling site during the NPF event periods, e.g., the mixing ratio of SO2 was < 3 ppb in the spring and < 5 ppb in the winter during the periods of NPF events. Less polluted or even clear air exactly meets equation 4 in the manuscript. The authors cannot find what problem in our approach is.

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.

Response: The authors would like to believe that the reviewer may capture a piece of information about air pollution in Beijing, but it is hard to say the piece reflecting the full picture. The technical term “polluted atmospheres” is probably not applicable for NPF events reported in this study.

The authors had no simultaneous measurements of other condensing species, such as H2SO4, HOM, etc., and had no way to discuss them. The authors agree that the SO2 concentration at the two sites indeed needs more interpretation. In revision, we will add “The sulfur content in the gasoline and diesel was limited <50 ppm at that years. The measured BC spikes were lower than 5 μg m-3 during the NPF periods. The maximum contribution of traffic-related SO2 at the street site was roughly estimated to be 1.3 ppb according to the results in our previous studies (Meng et al., 2015 a,b)”. In the wintertime, the ratio of traffic-derived SO2 to the observed values was less than 1/4 and the observed values were overwhelmingly contributed by domestic heating. The uncertainty by assuming SO2 at the street site same as the rooftop site should be minor in the wintertime and it should not affect our conclusion because the formation rates of new particles at the street site were increased by 3-5 times against the rooftop site in the wintertime. In the springtime, the contribution of traffic-related SO2 might significantly increase the mixing ratio of SO2 at the street site. However, the reduced NPF was observed at the street site. The possible underestimation of SO2 at the street site further solidified our analysis results, i.e., a strong scavenge effect at the street site likely existed and caused the reduced NPF.

The authors believe the turbulence dispersion to be more important than advection diffusion at the street site under a strong synoptic wind. The authors didn’t measure wind direction and wind speed at multiple street locations and the authors didn’t think one point observation of wind direction and wind speed can full reflect the complicated micro-scale wind field at the street site. The authors thereby are reluctantly speculated

the influence of the complicated micro-scale wind field on the observed particle number concentrations between the rooftop site and street site.

Through a deep data analysis, the authors additionally provided two types of unique evidences which were less affected by micro-meteorology at the street site as well as the simplest evidence, i.e., the simple comparison between the rooftop site and street sites, to confirm the reduced and enhanced NPF at the street site in different seasons. The authors found that the sufficiency and uniqueness of three types of evidences were not fully recognized by the reviewers because their challenges mainly focused on the simplest evidence. This means the authors' presentation strategy has to be improved to make the three types of evidences more obviously.

In revision, the authors will clarify that the reduced NPF always occurred at the street site in the springtime. Three evidences from different angles will be itemized as Evidence 1, 2 and 3, i.e., Evidence 1: The lower particle number concentration (PNC) of nucleation mode particles at the street site mainly because of a shorter initial burst time. Evidence 2: The authors used the PNC at the street site subtracting the corresponding PNC at the rooftop site to calculate the difference. The authors then obtained the second evidence: the negative difference of nucleation mode particles against the positive difference of Aitken mode particles on NPF days. Evidence 3: Using the same approach, the authors obtained the third evidence: the negative difference of nucleation mode particles on NPF days against the positive difference of that on non-NPF days (Figs. 3 and 4 in the origin version).

In addition, the authors will clarify that the enhanced NPF always occurred at the street site in the wintertime. Three evidences from different angles will be itemized as Evidence 1, 2 and 3, i.e., Evidence 1: The significantly larger PNC of nucleation mode particles at the street site and a larger apparent formation rate of new particles mainly because of a shorter initial burst time. Evidence 2: The positive difference of nucleation mode particles in the wintertime against the negative difference of nucleation mode particles in the springtime on NPF days. Evidence 3: The larger positive differ-

ence of nucleation mode particles on NPF days against that on non-NPF days in the wintertime (Figs. 5 and 7 in the origin version).

In this study, NOx was not measured at the street site. However, black carbon (BC) was measured by a potable aethalometer at the street site and BC is also a good indicator of traffic emission (Fruin, et al., 2004; Meng et al., 2015 a,b). The authors tried to use BC as an indicator of vehicle emission plumes to deduct primary traffic particles. It does not work because the one-minute time resolution is too low to successfully deduct primary traffic particles. To best of our knowledge, NOx analyzers are usually set for operating in one-minute time resolution and the data of NOx may suffer from the same problem. An example is presented to illustrate the problem for using BC to deduct primary traffic particles.

During the entire sampling period on 22 December 2010, BC shows no correlation with the nucleation mode PNC (shown in Fig. 1a). During a few short periods, the BC spikes appeared to be visibly consistent with the PNC spikes as shown in Fig. 1b. However, the correlation obtained was much poor, e.g., during the period of 10:30-12:30 (shown in Fig. 1c). This is not surprised because the aethalometer reported the instantaneous value of BC in one minute, but the vehicle spikes physically occurred in a few seconds (shown in Fig. 2). Under such poor correlation, the regression equation is invalid to accurately deduct primary traffic particles.

The points above justify a major reappraisal of the data, and the development of far less ambitious conclusions.

Response: The authors will try the best to revise to improve the manuscript. The authors have to say that "polluted atmospheres" in Beijing much claimed by the reviewer may be not closely related to NPF events reported in this study. Polluted atmospheres in Beijing indeed take place in presence of stagnant metrological conditions or under a dominant south or southwest wind.

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.

Response: Agree, the authors will revise the part as: “Urban street canyons provide semi-enclosed environments trapping vehicle exhausts that contain aromatic and aliphatic hydrocarbons, SO2, NOx, amines, black carbon, etc. (Pierson et al., 1983; Stemmler et al., 2005; Burgard et al., 2006; Liu et al., 2008; Buccolieri et al., 2009; Sun et al., 2012; Gentner, et al., 2012)”

(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 respiratory 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.

Response: The authors will revise as: “Ultrafine particles (<100 nm) have been reported to have adverse human health impacts through the deposition in the pulmonary region and penetration into the bloodstream (Oberdörster et al., 2004; Schlesinger et al., 2006; Zhang et al., 2012, 2015).” “In addition, the newly formed particles inside a street canyon may become toxic when vehicle-released organics is involved in the nucleation process (Sgro et al., 2009; Gualtieri et al., 2014).”

(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.

Response: As reported by Zimmerman et al. (2015), an independent measurement of CPC simultaneously with FMPS can be used to accurately correct the FMPS data including the size-dependent particle loss. The reference has been cited in the manuscript. In this study, a CPC was operating simultaneously with a FMPS at the street site and the FMPS was thereby used to correct the other. The two FMPS were

then processed the second-step correction proposed by Zimmerman et al. (2015). The total particle number concentration of the FMPS at the rooftop site was multiplied by a correction factor which was equal to the minutely averaged ratios of the FMPS and CPC data at the street site. The calculated value and the CPC data was used to complete the third correction for the FMPS data at the rooftop site and at the street site, respectively. This will be clarified in revision.

The particle loss for >8 nm particles were undetectable in 2.8 m sampling lines. This will also be added in the revision.

(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.

Response: The equation (3) in this study was exactly same as reported by Dal Maso et al. (2005) and Kulmala et al. (2005). The references have been cited in this study. The authors don’t understand the equation presented by Kulmala et al. (2012) and prefer to use the equation widely adopted in literature.

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

Response: “the net maximum increase of nucleation mode PNC (NMIoNP)” is calculated as “N8-20nm(t1)-N8-20nm(t0)”. t0 is set as the time when the apparent NPF started to be observed and t1 as the time when the nucleation mode PNC reaches the maximum value. This will be added in revision.

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

Response: In revision, the authors will reorganize the evidences to confirm Class II particles to be a regional NPF event, i.e.,

1) As reviewed by Vu et al. (2015), the particle number size distribution (PNSD) of emission source (e.g., traffic emissions, industrial emissions, biomass burning, cook-

ing) character the typical peak number mode, such as at 30 nm, 50 nm, 70-80 nm, 120-140 nm, et al. When the NPF events in Class II occurred in our study, the nucleation mode particles overwhelmed and other particle modes were negligible.

2) Similar to Class II NPF events with the particle growth to be undetectable presented in this study, extremely low growth rate of newly formed particles ($\sim$ 1 nm h-1) in Beijing was also previously reported by Wehner et al. (2004). In our unpublished data, the authors simultaneously observed Class II NPF events and NPF events with extremely low growth rate at $\sim$240 km distance (shown in Fig. 3, the case will also be presented in Supplementary). In the last three years (data unpublished), we had simultaneous observations of NPF events at 100-500 km distance. The authors obtained six cases based on simultaneous observations at two locations, i.e., one case featured by Class II NPF vs Class II NPF, four cases featured by Class II NPF vs NPF with an extremely low growth rate, one case featured by Class II NPF vs NPF with "banana shape" particle growth.

3) In this study, the duration period of Class II events lasted for 4-8 hours with the wind speed >4m/s. The authors strongly believed that they should be considered as regional NPF events.

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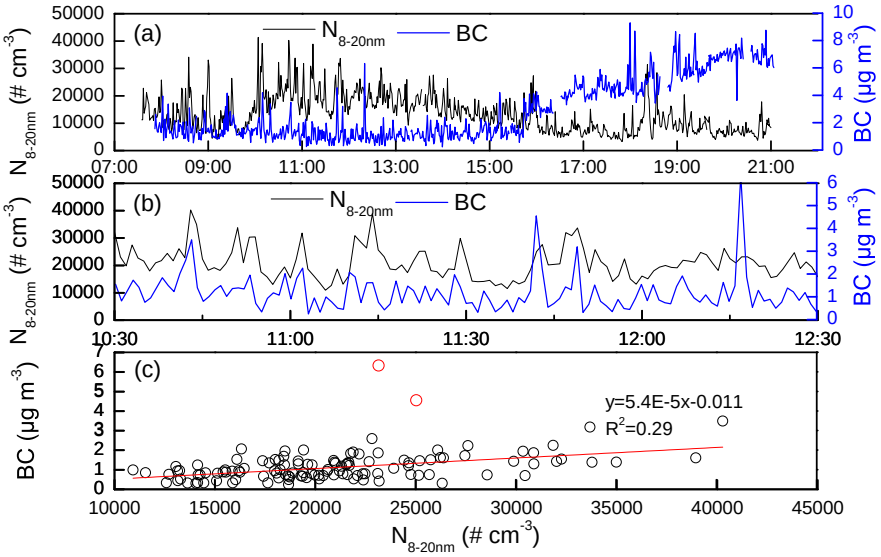

Fig. 1. The nucleation mode PNC and BC on 22 December, 2010. (a, b: time series of nucleation mode PNC and BC; c: relationship between nucleation mode PNC and BC during 10:30-12:30)

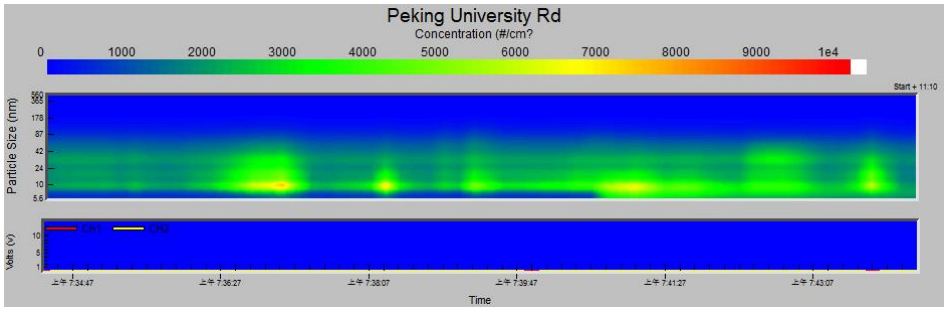

Fig. 2. Raw FMPS data showing vehicle spikes.

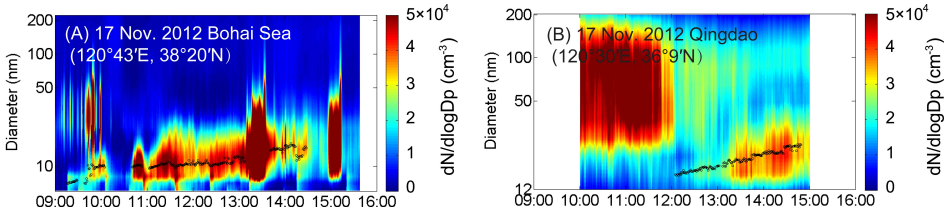

Fig. 3. Simultaneous observed Class II NPF event and NPF event with extremely low growth rate at ∼240 km distance.