Measurement Report: New particle formation characteristics at an urban and a mountain station in Northern China by Ying Zhou et al.

In this file, the referee comments are in black, our item-by-item replies are in blue, and the corresponding modifications in the manuscript are in red.

Answers to reviewer # 1

The authors extend the dataset and the paper has been improved significantly. However, there are some issues that the author should give more explanations. There are also some grammatical and spelling errors in manuscript. The authors need to check through the manuscript carefully.

We would like to thank the referee for their suggestions and careful editorial comments. We present our answers to the referee comments point-by-point below and paid careful attention to the grammatical and spelling errors in manuscript.

Major concerns:

1. The accumulation mode particles number was reported to be ~700 cm^{-3} on NPF days on urban site, which is almost 50% lower than the mountain site. The author should check the data, as it was also reported the CS on NPF days at both sites was ~0.01 s^{-1}. As the CS values are quite similar and dominated by the accumulation mode, why the difference in accumulation mode concentration between the two sites is so large? Also, it should be clear that the size range of each mode in the text.

We revisited our data and calculation of CS as well as particle number concentration of every mode at both sites.

As shown in Figure R2-1 (c&e) on NPF event days, particles smaller than 100 nm contribute to a CS of 3.7×10^{-3} s^{-1}, contributing 37% to the total CS. While at MT site, particles smaller than 100 nm only contribute to a CS of 1.2×10^{-3} s^{-1}, contributing less than 12% to the total CS (Figure R2-1 (d&f)). Although 100-840 nm particle number concentration at UB site was much less than that at MT site, 1-100 nm (especially 25-100 nm) particles compensated total CS by higher number concentration on NPF event days.

The nucleation, Aitken and accumulation mode particles we mentioned in our manuscript were particles in the size ranges of 6-25 nm, 25-100 nm and 100-840 nm, respectively.

As per suggestion by the reviewer, we updated Figure 6 in the manuscript as Figure R2-1 and add discussion on CS in line 264 as below:

As shown in Figure 6 (c&e) on NPF event days, particles smaller than 100 nm contribute to a CS of 3.7×10^{-3} s^{-1}, contributing 37% to the total CS. While at MT site, particles smaller than 100 nm only contribute to a CS of 1.2×10^{-3} s^{-1}, contributing less than 12% to the total CS (Figure 6 (d&f)). Although 100-840 nm particle number concentration at UB site was much less than that at MT site, the 1-100 nm (especially 25-100 nm) particles largely participated by higher number concentration on NPF event days to result in a comparable CS between both sites.

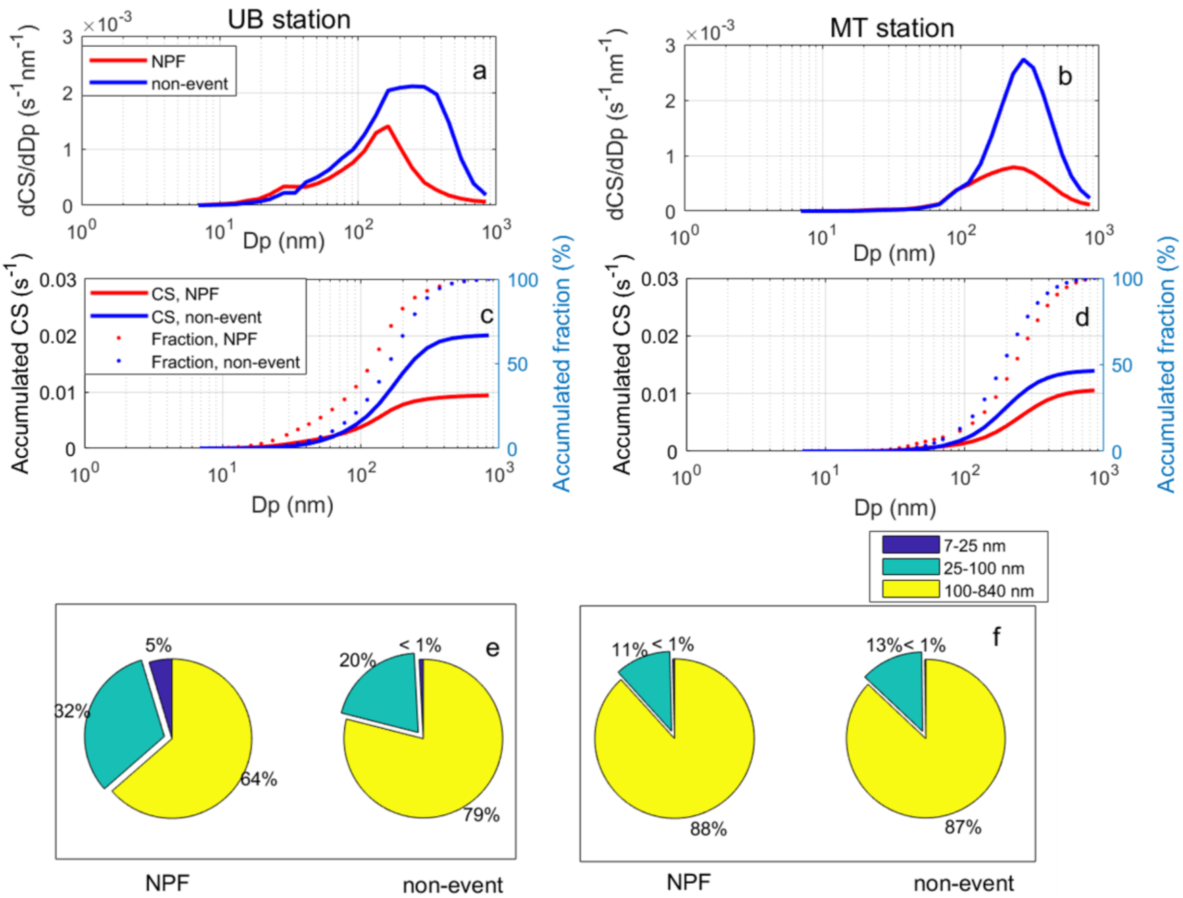

Figure R2-1: Median CS size distribution (a&b), accumulated CS contributed by particles from 6 nm and the ratio between accumulated CS and total CS (c&d); Contribution of size-segregated particles to total CS (e&f) at each site on NPF and non-event days during 9:00-15:00 (local time, LT). Figures on the left and right panels represented data observed at UB and MT site, respectively. The time resolutions for CS and particle number concentration data were 8 min at UB station and 4 min at MT station, respectively.

As per suggestion by the reviewer, we added size range of each mode in line 534-536 as below:

On NPF event days, nucleation (6-25 nm) and Aitken (25-100 nm) mode particle number concentrations were much smaller at MT station than those at UB station due to smaller formation rates and less anthropogenic emissions. Interestingly, accumulation (100-840 nm) mode particle number concentrations….

2. In the summer campaign, which is rainy season in Beijing, the precipitation should be addressed as it is an important scavenging process of particles. Also, for Mountain site, the fog/cloud process is another particle scavenging process that can influence the CS.

We thank the reviewer for the comments. In summer 2018 and 2019, the DMPS data were discarded from analysis on rainy days as rain affected the quality of the DMPS data. We calculated CS at both sites assuming RH equals to 0%. When we calculated the RH affected CS at MT site, e.g. 30%-70% during 9:00-15:00, as shown in Figure 10, the CS would be 1.12-1.33 times of those with a RH of 0%, as shown in Figure R2-2.

As per suggestion to the reviewer, we added the following discussion in line 264:

The data on rainy days were discarded from analysis at both sites, hence the precipitation was considered to have minor effects on our CS calculation. We calculated CS at both sites assuming RH as 0%. It should be noted that the CS may have been underestimated by a factor of 1.12-1.33 at MT site when we include RH in the CS calculation, e.g. 30%-70% during 9:00-15:00, as shown in Figure 10.

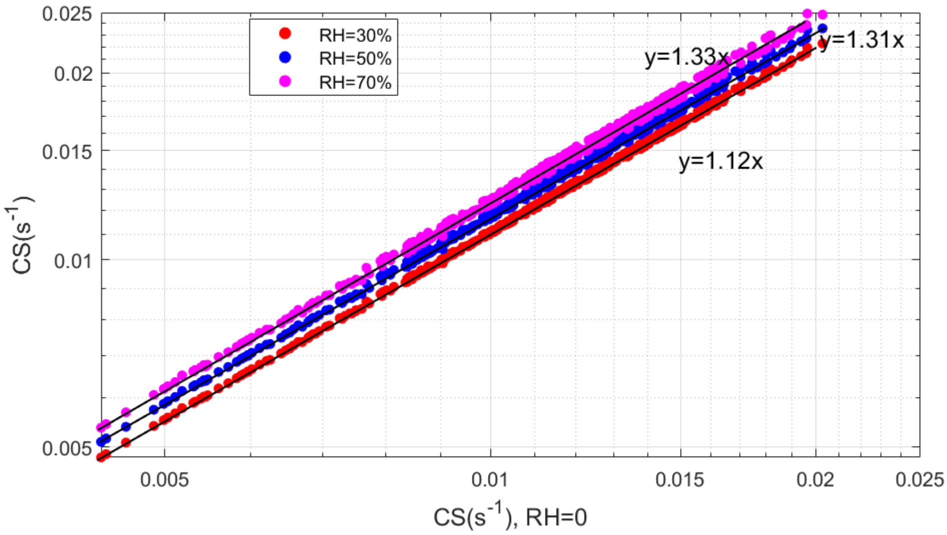

Figure R2-2: CS calculated with RH at MT site (30%, 50% and 70%) as a function of CS calculated with RH as 0%. The slopes of the fitted lines represent ratios between CS with RH at MT site and RH as 0%.

Specific comments:

1. Line 27, it's not necessary to give the reference in the abstract.

We removed the reference from the abstract.

2. Line 38-40, the author addressed the CS during the first two hours of NPF, indicating the concentration level of the pre-existing particles. However, why the formation rates during the first two hours of NPF are compared?

We thank the reviewer for the suggestions. Determination of nucleation start and stop times were difficult at UB site due to the contribution of traffic emissions. Hence, we choose a time window of the first 2 hours of NPF event for formation rates calculation at both sites for objectivity. During the time window, we always observed 7-10 nm particle number concentration burst significantly from the background level at both sites.

According to the comments of the reviewer, the following sentence will be added in our manuscript of line 286:

Determination of nucleation start and stop times was affected by traffic emissions at UB station. Hence, we choose a time window of the first 2 hours of NPF event for formation rates calculation at both sites. During the time window, we always observed 7-10 nm particle number concentration burst significantly from the background level at both sites.

3. Line 110, please shorten “particle formation rate, particle growth rate” to “formation rate and growth rate”. Check these words through the manuscript.

Corrected.

4. Line 125, the site S60 and LQ should be illustrated in the site description, not in the figure caption. In the discussion, S60 station is not mentioned.

We thank the reviewer for the suggestion. As suggested by both reviewers, the S60 site has been removed from the discussion as well as the map in our manuscript. The introduction of LQ station has been added to the method’s section as below:

Longquan station: The Longquan national monitoring station sits in Longquan town, Mengtougou District, Beijing. It is 20 km west to UB site and 60 km east to MT site and considered a suburban station. The location is referred to as ‘**LQ**’ from here after and is shown on the map in Figure 1.

5. Line 138, it should be Salma et al., (2011)

Corrected.

6. Line 156-157, PNSD measured by PSD and DMPS matched with a factor of 2…it is not clear which PNSD is higher? The ratio of 2 is derived by the total number concentration or what else? In Fig.3, it seems the PSD is higher below 20 nm and 300-600 nm, but lower above 600 nm. Which data is referred as the true value?

We thank the reviewer for the comments. We chose PSD data as reference, e.g. PSD data is referred as true value. The ratio of 2 is derived by particle number size distribution. As per suggestion by the reviewer, we corrected the sentences in Line 156-157 as following:

The PSD was used as reference. As shown in Figure 3, particle number size distribution measured by DMPS matched well with PSD in data trend. Varying with particle diameter, particle number size distribution data measured by DMPS can be higher or lower than PSD within a factor of 2.

7. Line346, As shown in Figure ?? which figure? Line 351, in Figure 8a. The initial letter of Table and Figure should be capitalized. Please check all through the manuscript. Figure and

Fig are both used, it should be consistent.

We checked all through the manuscript for consistency and corrected Line 346:

As shown in Figure 8a,…

As per suggestion by the reviewer, we used Figure in our manuscript and changed all “Fig” into “Figure”.

8. Section 3.4 I suggest to revise the end Dp as *D*p,end, also be consistent with figure caption.

We thank the reviewer for the suggestions.

To avoid ambiguity, we changed the figure caption in the manuscript as following:

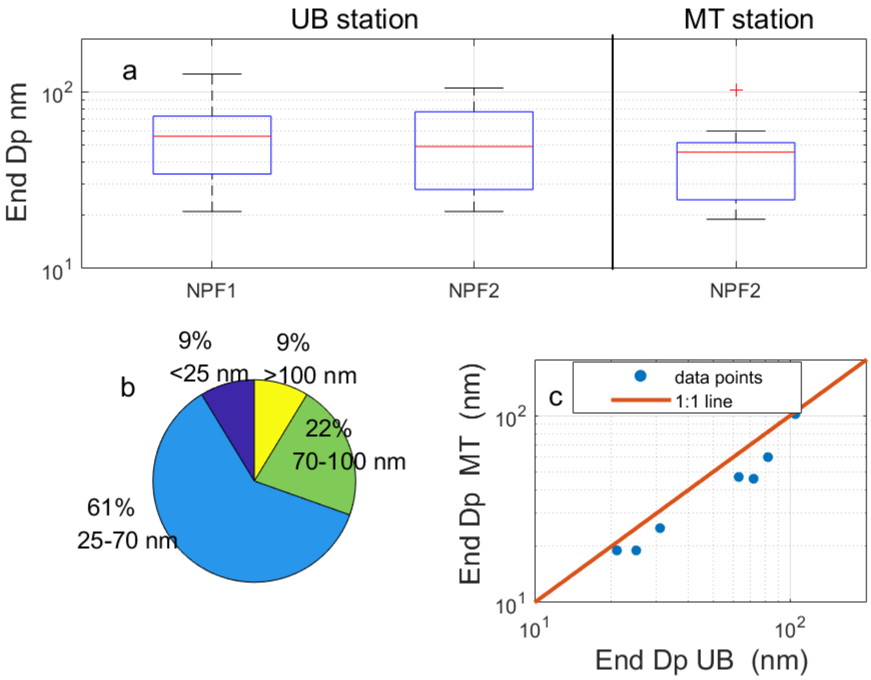

Figure 12: (a) Median and percentiles of end diameters (End Dp, nm) of NPF events measured at both sites. The red line represents the median of the data and the lower and upper edges of the box represent 25th and 75th percentiles of the data, respectively. The length of the whiskers represents 1.5× interquartile range which includes 99.3% of the data. The ‘NPF1’ and ‘non-event1’ referred to NPF event and non-event days in summer 2018 and 2019 and the ‘NPF2’ and ‘non-event2’ referred to NPF event and non-event days during the observation from June 14 to July 14, 2019. (b) Frequencies of end diameters in the size range of smaller than 25 nm, 25-70 nm, 70-100 nm and above 100 nm during our observation at UB

station in summer 2018 and 2019. (c) Comparison between end diameters of coincident NPF events at both stations.

9. I suggested each subplot in Fig.14 and other figure panels should be marked as a, b, c, d,…, in order to be referred easily. Please also check the figure captions as some are not complete, e.g., Fig. 18, the marker of mode diameters is not given.

We thank the reviewer for the comments. As per suggestion by the reviewer, we corrected Figure 14 as bellow.

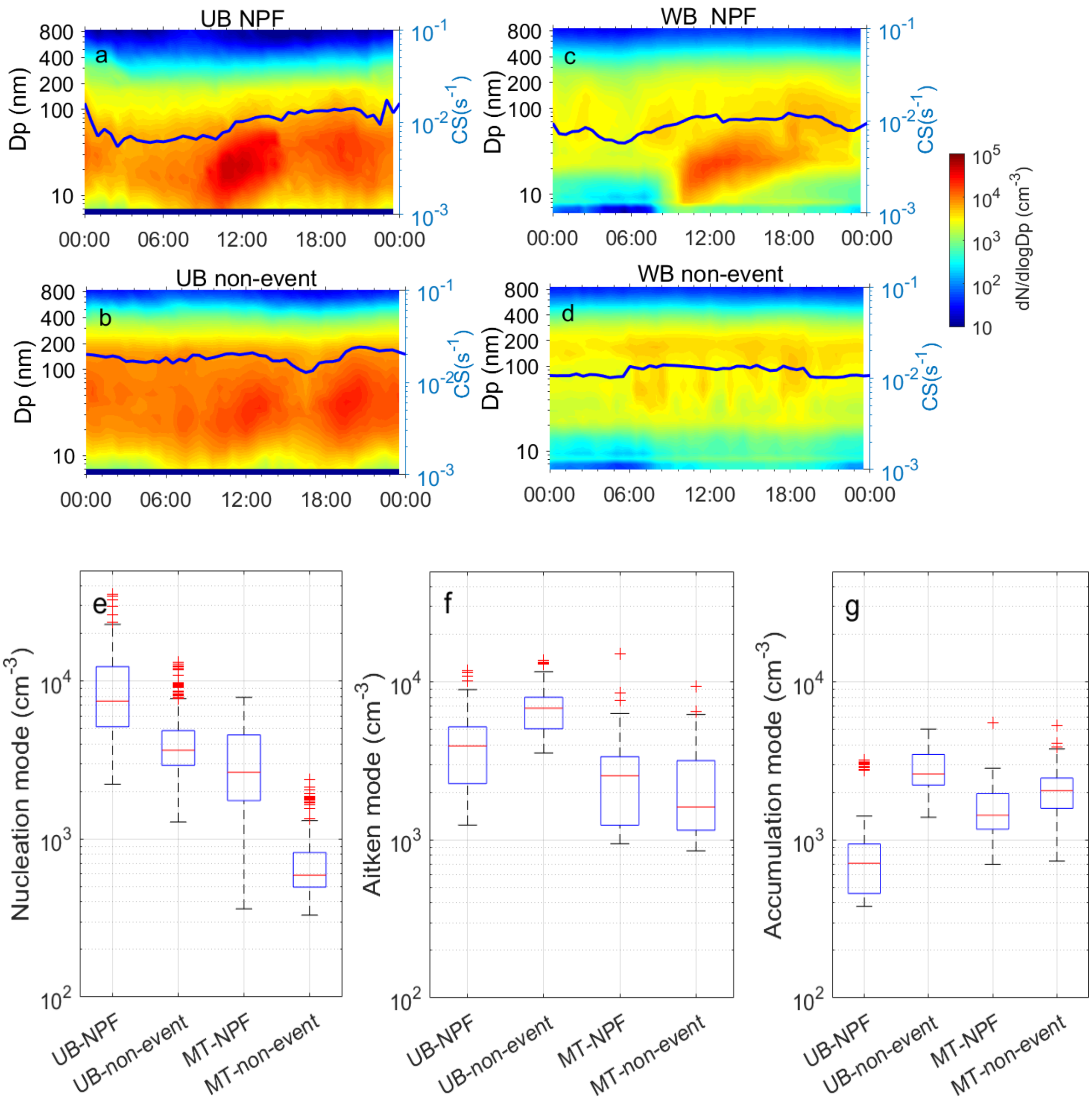

Figure 14: Median particle number size distribution as well as CS (blue lines) on NPF event and non-event days at UB (a &b) and MT (c&d) stations and median and percentiles of

nucleation (e), Aitken (f) and accumulation (g) modes particle number concentration on NPF event and non-event days during our observation from June 14 to July 14, 2019 at both stations. The red line represents the median of the data and the lower and upper edges of the box represent 25th and 75th percentiles of the data, respectively. The length of the whiskers represents 1.5× interquartile range which includes 99.3% of the data. Data outside the whiskers are considered outliers and are marked with red crosses.

As suggested by another reviewer, we removed section 3.6 and Figure 18.

10. The value of CS is suggested to be added in Table 1.

We added values of CS in Table 1 as follows:

Table 1: NPF event and non- event days during our observation at both stations.

Date	Type	Air masses (9:00-15:00)		$GR_{7\text{-}15nm}$ (nm/h)		J_7 ($cm^{-3}s^{-1}$)		Event Start (LT)		Ending diameter (nm)		CS (s^{-1})	
		UB	MT	UB	MT	UB	MT	UB	MT	UB	MT	UB	MT
2019/06/14	a	North	North	8.61	-	4.97	-	9:00	8:00	71	-	0.017	0.008
2019/06/15*	a	Local	Local	12.63	-	5.56	-	11:00	15:00	82	60	0.013	0.029
2019/06/17	d	East	Local									0.031	0.011
2019/06/18	c	Local	West		10.5		0.17		12:00		45	0.039	0.008
2019/06/19	d	South	Local									0.037	0.047
2019/06/21	d	East	Local									0.035	0.018
2019/06/23	e	East	East									0.033	0.013
2019/06/24	f	Local	Local		8.21		-		12:00		50	0.027	0.014
2019/06/25*	a	Local	Local	-	-	-	-	12:00	15:00	-	53	0.032	0.027
2019/06/28	g	West	West	-		-		11:00				0.022	0.006
2019/06/29	a	North	North	12.93	7.14	6.93	2.28	9:00	8:00	21	19	0.008	0.011
2019/06/30	a	North	North	4.82	6.57	9.86	1.37	6:30	9:30	31	25	0.003	0.008
2019/07/01	a	North	North	7.31	5.82	3.84	0.82	9:00	8:30	105	102	0.006	0.009
2019/07/02	d	Local	West									0.013	0.014
2019/07/03	a	North	North	7.89	6.52	3.25	0.75	9:00	8:00	72	46	0.015	0.006
2019/07/04	b	Local	Local	-		-		10:00		53		0.012	0.012
2019/07/06	a	North	North	7.39	6.51	9.21	1.75	7:00	9:30	25	19	0.004	0.011
2019/07/07	b	North	North	7.61		3.61		9:00		32		0.008	0.005
2019/07/08	d	East	East									0.019	0.012
2019/07/09	d	East	East									0.021	0.015

2019/07/10	h	East	East									0.017	0.013
2019/07/11	d	East	East									0.039	0.014
2019/07/12	f	East	East		5.57		0.37		9:30		24	0.018	0.014
2019/07/13	c	Local	North		6.32		0.70		10:00		30	0.037	0.012
2019/07/14	a	North	North	12.04	9.86	3.91	0.89	9:30	9:30	63	47	0.023	0.017

'a' means NPF event observed at both stations, 'b' means NPF event day at UB station while non-event day at MT station, 'c' means NPF event day at MT station while non-event day at UB station, 'd' means non-event day at both stations on the same day, 'e' means undefined day at both stations, 'f' means undefined day at UB station while NPF event day at MT station, g means undefined day at MT station while NPF event day at UB station, h means undefined day at UB station while non-event day at MT station, * means NPF event observed at MT station was transported from somewhere else. – means the values cannot be reliably calculated. Only days when particle number size distribution were valid are included in this table.

11. Spelling check (including the below, but not limited to):

Line 112, “favorable conditions”;

Corrected

Line 114, conditions those… could help to minimize…

Corrected

Line 295, NPF characteristics;

Corrected

Line 239, said times??

We modified the sentence as “NPF event start and stop times” in our manuscript.

Line 388, is considered to be one of the most…

Corrected

Line 511, please check Fig. R12c

Corrected

Line 568, This is a common

Corrected

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Answers to reviewer # 2

This work reported simultaneous measurements of new particle formation events during an intensive campaign at an urban and a mountain station in China. It is a complex and extended study that fits well with to the scope of ACP and it is of interest for the international research community. However, there are some issues to be improved or corrected before it is published in ACP.

We would like to thank the referee for the suggestions and careful editorial comments.

We present our answers to the referee comments point-by-point below.

<u>Major comments</u>

1. The authors include a long paragraph in the introduction to summarize some results on the regional extension of NPF. This manuscript deals with the extension of NPF events but also with two different altitude sites (one mountain site). In my opinion, it is necessary to include a paragraph about the state-of-the-art of NPF events at mountain sites and the vertical distribution of NPF. Sellegri et al (2019) reviewed NPF events at mountain sites and it is not cited along the manuscript. This same manuscript also discusses the topography or preferred altitude of NPF events. In addition, there is some studies that attempt to look on the vertical distribution of NPF (e.g., Komppula et al., 2003; Boulon et al., 2011). Finally, there is also a recent and similar study that looks on the differences of NPF at two sites, also urban and mountain sites, (Casquero-Vera et al., 2020) and the differences or similarities of the results should be discussed with this similar study where the GR, J or CS are also discussed at two different altitude sites.

We thank the reviewer for the suggestions. We added a paragraph between line 104 and line

105 in our manuscript as below:

In addition to horizontal extension of NPF events, the vertical extension of NPF events also attracts attention of researches. It have been confirmed that NPF events can be triggered within the whole low tropospheric column at the same time and even above the planetary boundary layer upper limit (Boulon et al., 2011). Sellegri et al. (2019) reviewed NPF events observed at 6 different altitude stations. They found NPF events were most favored at the altitude close to the interface of the free troposphere (FT) with the planetary boundary layer (PBL) and at the vicinity with clouds. In addition, at high altitude sites, CS may not be the liming factor for NPF occurrence as higher CS associated with more precursors for nucleation and initial growth. Based on observations at two different altitudes (e.g. 340 m and 560 m above sea level) in northern Finland, Komppula et al. (2003) found NPF events had similar formation and growth rates between these two heights, while due to vertical movement of air masses, difference of NPF event start time between these two sites was limited within 30 min. Similar results were also observed at two sites in France that formation and growth rates were similar between two altitudes (e.g., 660 m and 1465 m above sea level) while the contribution of ion-induced nucleation was higher at high altitude (Boulon et al., 2011). Finally, during a recent observation in Spain, growth rates were higher at the mountain site (2500 m a.s.l.) than urban site (680 m a.s.l.), while difference between formation rates varied with altitude (Casquero-Vera et al., 2020).

2. In P19 the authors stated that urban emissions affect the formation rates, but the NPF are of regional extension? Could local events happen without that regional phenomena? Could the emissions of that huge city be the unique responsible of the regional NPF? In this same section, the authors suggest that "precursors needed for particle formation were much more abundant in the polluted urban environment (Wang et al., 2013), while those needed for growth are rather comparable". The analysis of J is for 7nm size, that means these particles are not newly formed, these particles come from growth or could be emitted directly by i.e. traffic? At MT, 7 nm particles means that these particles could not be formed there or the vicinity? Please clarify these ideas.

We thank the reviewer for the comments. As we discussed in section 3.4, the upwind extension of regional NPF events was limited to the areas with some anthropogenic emissions. There should not be any discrete boundary between the regions that NPF event is or is not occurring, but with decreasing anthropogenic emissions, the strength (formation rates and growth rates) are expected to decrease. Particle formation rates were usually positively correlated with H_2SO_4 concentration, the urban emissions can provide abundant SO_2, hence abundant H_2SO_4, resulting in high formation rate (Kerminen et al., 2018).

Local events can happen without regional phenomena. We sometimes observed nucleation mode particle number concentration burst without mode diameter increasing. It could be related to non-regional NPF events (Dai et al., 2017). We did not observe such cases at the MT site. Actually, the abundant anthropogenic emissions in the megacity could provide enough precursors for non-regional NPF events. However, traffic emissions can also provide abundant primary nucleation mode particles making it difficult to distinguish whether the new mode was from NPF event or traffic. So we classified such events as “undefined” also.

During our observation, there was no air mass convection between two sites. And we did not conduct chemistry measurement at MT site, as a result, the contribution of urban emissions on particle growth is unknown there. So we are not sure whether urban emissions can be the unique responsible of regional NPF events. To figure this question out, we still need long time observation on gas and particle phase chemistry as well as particle number size distribution down to sub-3 nm downwind urban Beijing.

Due to the non-zero wind conditions, the 7 nm particles we observed at both sites should have originated upwind the sites. In addition, Boulon et al. (2011) observed that new particles could be formed at low altitude and transported to the higher altitude sites. However, to confirm whether the phenomenon can happen at MT site, we still need observations on vertical wind conditions or vertical evolution of potential temperature. At UB site, the traffic emissions can also provide 7 nm particles, but compared with NPF events, the contribution of traffic emissions is considered minor (Zhou et al., 2020).

As per suggestion by the reviewer, we updated our conclusion in line 640 as below:

The upwind extension of regional NPF events was limited to the areas with some anthropogenic emissions. There should not be any discrete boundary between the regions that NPF event is or is not occurring, but with decreasing anthropogenic emissions, the strength (formation rates and growth rates) should decrease.

As per suggestion by the reviewer, we added such sentence below in line 251 as below:

At UB site, we also observed some cases in which nucleation mode particle number concentration burst with mode diameter increase. It could be related to non-regional NPF events (Dai et al., 2017). We did not observe such cases at the MT site. Actually, the abundant anthropogenic emissions in the megacity could provide enough precursors for non-regional NPF events. However, traffic emissions can also provide abundant primary nucleation mode particles, making it difficult to distinguish whether the new mode was from NPF event or traffic. So we classified such events as “undefined” also.

As per suggestion by the reviewer, we updated our conclusion in line 644 as below:

For more robust knowledge on NPF events in north China plain and to figure out the effect of urban emissions on regional NPF events, we still need long-term observations including particle number size distribution down to sub-3 nm, gas and particle phase chemistry downwind and upwind urban Beijing.

As per suggestion by the reviewer, we updated our conclusion in line 644 as below:

Also, the J_7 at UB station could be affected by traffic emissions due to the proximity of the location to the highway, while compared with NPF events, the effect of traffic emissions is shown to be minor (Kontkanen et al., 2020; Zhou at al., 2020). In addition, Boulon et al. (2011) observed that new particles could be formed at low altitude and transported to the higher altitude sites, however, to confirm whether the phenomenon can happen at MT site, we still need observation on vertical wind conditions or vertical evolution of potential temperature.

3. Case studies are “special cases” but, in my opinion, they are not analyzed in depth. For example, shrinkage cases are of interest since there is not clear the origin of this phenomenon (e.g., Salma et al., 2016; Alonso-Blanco et al., 2017). In this sense, Section 3.6.3 makes an

attempt to discuss the case of stagnant and shrinkage but unfortunately, there is not a real study or discussion of this special case. Please go further on this or remove these sections.

As per suggestion by the reviewer. With the data we have now, we are not able to provide much clarification on mode diameter shrinkage issue. So we removed the case studies part, e.g. section 3.6.

4. Please review the whole text, there is long sentences without any comma and not well connected along the manuscript.

As suggested by both reviewers, we have corrected all the grammatical and spelling errors in manuscript as much as we can find.

Minor comments

L26 – Change "few" to "low"

Corrected.

L41 – "at urban site" is repeated

Corrected.

L128 – And the altitude of the urban site?

We thank the reviewer for the suggestion. The altitude of the west campus of BUCT is around 50 m above sea level and the urban site is located on the fifth floor of a university building inside the west campus of BUCT, around 12 m above ground level.

According to the comments of the reviewer, the following sentence will be added in our manuscript of line 122:

The altitude of the west campus of BUCT is around 20 m above ground level and the urban site is around 12 m above ground level.

L138 – Cite format

Corrected.

L148-149 – Why do you mention Fig 2 here? Strictly, both instruments cannot correlate if they don't measure at same time.

We thank the reviewer for the suggestion. We mention Figure 2 here to show a general data quality of particle number size distribution during our short time parallel observation. The

FMPS matched well with SMPS in laboratory comparison after being well calibrated.

According to the comments of the reviewer, we removed the sentence in line 147 and add the following sentence below in line 134:

As shown in Figure 2, the data qualities of particle number size distribution at both sites during the short-term parallel observations was good in general.

According to the comments of the reviewer, we modified the sentence in line 148 in our manuscript as below:

The particle number size distribution measured by FMPS matched well with SMPS during the comparison in laboratory after being calibrated.

L160 – It “is” reasonable

Corrected.

L164 – What calibrations were done?

The particle number size distribution from FMPS were calibrated according to the method introduced by Zimmerman et al. (2015).

According to the comments of the reviewer, we modified the sentence in line 164 in our manuscript as below:

On the other hand, the particle number size distribution from FMPS was carefully calibrated and the FMPS was properly operated during the observation as we discussed above.

L181 – Space after dot

Corrected.

L221 – must have been?

We thank the reviewer. We corrected the sentence as following:

This is because as time progresses, the particles observed at a measurement site should have originated from further and further away due to non-zero wind conditions.

L278 – Is it correct that the “dk” is included and the “du” is not? “[dk, du)”

We thank the reviewer for the comments. $N_{[dk,du)}$ is defined as the total number concentration of particles in the size range from dk to du (particles with diameters of du are not accounted for) (Cai and Jiang, 2017).

According to the comment of the reviewer, we corrected L278 in our manuscript as below:

$N_{[dk,du)}$ is defined as the total number concentration of particles in the size range from dk to du (particles with diameters of du are not accounted for).

L288 – NPF event "frequencies" is not an adequate title for this section. Maybe something on the "occurrence" but not the frequency. I suggest "Origin of NPF events at both sites"?

We thank the reviewer for the good suggestion. According to the suggestion by the reviewer, we changed the title of this section as "Origin of NPF events at both sites".

L291-293 – Rephrase

We thank the reviewer for the suggestions. We rephrased the sentence in our manuscript as following:

The NPF event frequency was consistent with an earlier observation in summer in urban Beijing from 2004 to 2008, while smaller than other seasons especially winter during that observation and another one-year observation in UB station.

L300-303 – Rephrase, use comma.

We thank the reviewer for the suggestions. We rephrased the sentence in our manuscript as following:

Data were considered as valid when visual inspection of the particle number size distribution data and the instrument status did not indicate problems in the measurements. Only days with valid data at both stations were taken into consideration in our analysis.

L304 – "Common" is referred to "coincident" events? If it is, change the terminology.

We thank the reviewer for the suggestions. We changed "common events" into "coincident events" in our manuscript.

L615 – Mechanisms are not really investigated

We thank the reviewer for the comments.

We deleted "and mechanism" in line 615 in our manuscript.

References

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