Review of Casquero-Vera et al. "New particle formation at urban and high-altitude remote sites in the south-eastern Iberian Peninsula" by Anonymous Referee #1

We thank the reviewer for his/her valuable comments and suggestions that helped us to improve the quality of the manuscript. Our responses to the reviewer's comments are detailed below. Our answers to reviewer are shown in bold and the changes inserted in the manuscript are noted here in italic and between quotation marks. The changes in the new version of the manuscript are noted in red.

Response to referee #1:

This manuscript attempts to study NPF events at two contrastive sites, i.e., urban and high-altitude remote sites within 20 km distance. The authors found that NPF was associated with the transport of gaseous precursors from lower altitudes, always observed from the smallest measured sizes and had a higher growth rate of newly formed particles at the high-altitude site. They also analyzed the contribution of sulfuric acid in particle growth, the importance of CS and availability of VOC in NPF events. In my opinion, the paper is generally well-written and suitable for publishing in ACP. A few minor comments are listed for the authors considering.

A point by point response is included below.

1) Page 3, lines 3-10, the authors are encouraged to cite the papers, in which the contributions of NPF to CCN at specific supersaturations were measured directly.

Following the reviewer's suggestions, we included papers in which the contribution of NPF to CCN were measured directly and made the following changes:

"Typically, the size at which aerosols activate as CCN ranges from 50 to 150 nm (Kerminen et al., 2012). However, some observational studies have showed that particles do not have to grow to sizes above 50 nm to be able to act as CCN and particles as small as 20 nm can form cloud droplets (e.g., Fan et al., 2018; Leaitch et al., 2016; Leng et al., 2014). In this sense, NPF events are one of the main processes producing aerosols in these sizes that has been estimated to enhance CCN number concentration by a factor of 1.2-1.8, depending on supersaturation (e.g., Dameto de España et al., 2017; Leng et al., 2014)."

2) Page 7, the bottom paragraph and Page 8, the top paragraph, the reviewer gets lost by the comparison presented here. The similarity and difference between the contrastive sites in literature should be compared with the findings in this study, correct?

We agree with the reviewer that the comparison presented in the first paragraph of section 3.1 was not clear and different size ranges were used in this comparison. In order to improve this section, we made the following changes in the new manuscript version:

"In general, the direct comparison of the results obtained in this study with those reported in literature is difficult and large differences in the aerosol number concentrations between the different sites may result from differences in the measured size ranges, instrumentation, sampling period, site location and proximity to the sources. However, the average aerosol concentration measured at UGR urban station was in the range of those obtained in summer season (4-24 × 10^3 cm^{-3}) in other European urban sites (e.g., Birmili et al., 2016; Gómez-Moreno et al., 2011; Pérez et al., 2010; Pey et al., 2010). On the other hand, the mean aerosol concentration at SNS was slightly higher than that observed at Puy de Dôme (research station located at 1465 m a.s.l. in central France), where $N_{10-1000}$ were 2.5 × 10^3 cm^{-3} in summer (Venzac et al., 2009). However, the mean aerosol concentrations at SNS was significantly higher than that reported (mean $N_{10-1000}$ of 767 cm^{-3} in July) for Jungfraujoch (high-alpine station located at 3580 m a.s.l. in the central Swiss Alps) by Weingartner et al. (1999). Aerosol number concentrations at remote sites are rather influenced by long-range transport or/and by transport from lower altitudes in the case of remote mountain sites. Therefore, aerosol transport from lower altitudes (i.e. from Granada city) could partly explain the high aerosol concentration observed over SNS station."

3) Page 9, lines13-15, "For many years, it was thought that NPF events cannot take place in heavily polluted urban areas, since the high condensation sink (high pre-existing aerosol concentration) in these areas was considered detrimental in suppressing the formation and growth of particles', What situation is "heavily polluted", please add a quantitative definition.

We agree with the reviewer that heavily polluted is not a quantitative definition and we removed this term along the manuscript. We made the following changes in the new manuscript version:

"For many years, it was thought that NPF events cannot take place in urban areas, where the high pre-existing aerosol concentrations was considered detrimental in suppressing the formation and growth of particles due to high condensation sink."

4) Page 10, lines 28-29, "This fact could have a special importance on cloud formations, since larger GR at SNS mountain station could be translated to larger survival probability of NPF particles to reach CCN sizes, due to shorter time needed for

the growth." This is not necessarily true by considering the ceiling of particle growth from 10 nm to CCN size or even particle shrinkage, e.g., Man et al., EST, âA˘ R 49, â ´ A˘ R´ 7170-7178,âA˘ R JUN 16 2015.

We agree with the reviewer that larger GR does not necessarily mean that NPF particles will reach CCN sizes and that ceiling or shrinkage should be considered as limiting factors. In our case, ceiling size varies from 25 to 60 nm during class I events at SNS site and no shrinkage process was observed. That means that NPF events at SNS site will affect to CCN concentrations at least at high SS. Thus, we included the considerations made by the reviewer as follow.

"This fact could have a special importance on cloud formations, since larger GR at SNS mountain station could be translated to larger survival probability of NPF particles to reach CCN sizes, due to shorter time needed for the growth. However, this fact is of importance after considering that ceiling size and shrinkage are not limiting factors to reach CCN size. Since ceiling of particle growth mode ranges from 25 to 60 nm with a mean value of 41 nm during class I events at SNS site and shrinkage has not been observed at this site, NPF events at SNS site can affect CCN concentrations at least at high SS (e.g., Fan et al., 2018; Leaitch et al., 2016; Leng et al., 2014)."

5) Section 3.3, the aerosol acidity and aerosol phase state may also affect the growth rate of newly formed particles, please add the analysis if possible.

We agree with the reviewer that aerosol acidity and aerosol phase state affect the growth rate of newly formed particles but we can not estimate the evaluate of these mechanisms in our sites with the current available measurements. In this sense, we included in section 3.3 these mechanisms as possible processes involved on the observed differences.

"Also, as Boulon et al. (2011) and Manninen et al. (2010) pointed, ion-mediated nucleation could be promoted at higher altitudes compared to low altitudes, and therefore can contribute to the observed differences. However, as Lehtipalo et al. (2016) showed, this mechanism only accelerates the growth of newly formed particles at low concentrations of base compounds. When a strongly basic compound is present, the growth of newly formed particles can be greatly enhanced by acid-base clusters. Thus, the presence of stabilizing vapours could also be responsible of the differences observed."

6) Page 13, lines 15-24, the reviewer has the same concern as presented comment 3.

To avoid confusion, we removed lines 7-9 of P14 in the new manuscript version.

7) Section 3.5, no direct measurements of sulfuric acid vapor are one of major weaknesses here, and the weakness should be added.

We agree no direct measurements of sulfuric acid is one weakness in this work. In section 2.2 (Data analysis section) of ACPD version we already mentioned that: "*The choice of k value will have an influence on the absolute value of the H_2SO_4 concentrations, but not on the relative variability*", **and we also made the following changes in the new version of Results section (P15, lines 27-31 on new manuscript version):**

"The GR_{7-25} shows similar values on 26 June (6.0 nm h^{-1}) and 24 June (6.3 nm h^{-1}). This is contrary to the expected from CS and H_2SO_4 concentrations. Firstly, on 26 June the CS mean value was 70% lower than on 24 June. Secondly, despite no direct measurements of sulfuric acid were available, the sulfuric acid concentration estimated by proxy on 26 June (4.0×10^7 molec cm^{-3}) was 3 times higher than on 24 June (1.3×10^7 molec cm^{-3}). As discussed before, the estimated concentrations of sulfuric acid could only explain less than 10% of the observed GRs."

References

- Leaitch, et al., 2016: Effects of 20–100 nm particles on liquid clouds in the clean summertime Arctic.

- Leng et al., 2014: Impacts of new particle formation on aerosol cloud condensation nuclei (CCN) activity in Shanghai: case study.

- Fan et al., 2018: Substantial convection and precipitation enhancements by ultrafine aerosol particles.

- Dameto de España et al., 2017: Long-term quantitative field study of New Particle Formation (NPF)events as a source of Cloud Condensation Nuclei (CCN) in the urban background of Vienna.

- Lehtipalo et al. 2016: The effect of acid–base clustering and ions on the growth of atmospheric nano-particles.