Response to Reviewer #1:

This is a very comprehensive and carefully-made analysis on the influence of atmospheric new particle formation (NPF) on CCN concentrations and eventually on cloud droplet number concentrations (N) in the Mediterranean atmosphere. The authors introduce a new approach to estimate CCN production from NPF, and then use model simulations to get N. The paper is scientifically sound and well written. There a few incorrect statements in the paper, and a few places that require further discussion. I consider, however, these issues minor, since they do not require major effort or changes in writing of the text.

The authors would like to thank the reviewer for the positive feedback and thoughtful and thorough comments. A point-by-point reply to all of the issues raised and the corresponding changes follows.

Important scientific issues:

One main findings stressed by the authors is the suppressed effect of NPF on cloud droplet number concentration because the maximum supersaturation reached in a cloud updraft is lower at higher CCN concentrations. There are at least two things related to this point that should be discussed, or at least mentioned briefly, in the paper:

First, the non-linear response of the cloud droplet number concentration (N) to the CCN concentration, or to any bulk property representing the amount of aerosol particles, is a well-known feature reported in a number of model studies investigating cloud droplet activation, as well as in several field measurements.

We completely agree! These points will be further clarified in the text, adding representative references over decades (e.g. Twomey et al., 1977; Ghan et al., 1993; Boucher and Lohmann, 1995; Gultepe, and Isaac, 1996; Nenes et al., 2001; Ramanathan et al., 2001; Ghan et al., 2011; Sullivan et al., 2016) that express this sub-linearity.

Second, practically all cloud properties (albedo, probability of rain formation etc.) are expected to become more or less saturate at high concentrations of CCN (to some extend also at high N). This means an increase of the CCN concentration by a

certain factor matters more in cleaner air. Since in most environments NPF is favored by low pre-existing particle concentration (i.e. cleaner air), this further means that the influence of NPF on cloud properties is usually expected to be greater than the influence of primary particle pollution in dirtier air.

The point is well taken. In the manuscript we make very clear that everything discussed refers to the conditions seen at the eastern Mediterranean, and that the background values prior to the NPF event determine the extent to which the additional CCN can affect cloud properties. These points will be further emphasized in the conclusions and abstract of the revised paper.

In response to the comments raised, we looked into the Finokalia datasets among the 162 days with NPF episodes and note the relationship between the pre-existing particle concentration and the CCN enhancement. Indeed, the increase of the CCN concentrations when NPF recorded under cleaner air conditions is slightly higher (on average ~7.5±5.3%) than in polluted days (on average 7.0±4.9%) and will be noted in the discussion and conclusions as well.

Third, the authors correctly point out the assuming a constant cloud supersaturation biases the estimated influence of CCN (and hence NPF) on N. However, they come to this conclusion by assuming a constant cloud updraft velocity w (or its dispersion). The magnitude of w certainly depends on environmental conditions. This means, for example, that while it is not fair to assume a constant cloud supersaturation, it may also not to be fair to compare different seasons by assuming the same w at every season.

We understand the reviewer's point. In the approach adopted here, the method itself could use actual observations of vertical velocity (w). This is not done here, because measurements of in-cloud vertical velocity were not available to use. That said, *a*) we use the method of "characteristic velocity" (Morales and Nenes, 2010), so that the droplet numbers calculate correspond to the average value over the probability distribution of positive vertical velocity; *b*) the marine boundary layer does not exhibit a considerably wider spectral dispersion higher than 0.3 m s^{-1} ("base-case" characteristic velocity, reflecting summertime boundary layer conditions in the vicinity of Finokalia), especially when considering the sensitivity test, where the vertical velocity dispersion was doubled (σ_w=0.6 m s^{-1}). The above-mentioned distribution of vertical velocity in the marine boundary layer, is consistent with vertical velocities observed in marine boundary layers (e.g. Albrecht et al., 1998 and references therein; Fountoukis et al., 2007; Ghan et al., 2011), where w displays a spectral dispersion around zero value (σ_w is calculated to be between 0.2 and 0.3 m s^{-1}); *c*) the relative contribution of NPF to CDNC was similar between these cases (21.9±6.5%,). Given the above, and considering that seasonal changes in σ_w do not exceed the range examined, we feel that the results presented here provide a fair representation of the seasonality of NPF effects on CDNC.

The authors estimate that NPF contributes to 39-69% of the CCN budget in the supersaturation range 0.38-1%. It should be noted their approach (as all the available approached based on field measurements) is only able to count on the influence of NPF on CCN if the newly-formed particle reach CCN size within less than a day or so after NPF. It is very likely that there are newly-formed particles that grow slower and still survive to become CCN later on. So, the real contribution of NPF to the CCN budget is likely to be somewhat higher than the numbers obtained from this analysis. This issue is worth to be mentioned in the paper.

We thank the reviewer for pointing out this important issue. We have mentioned this point early in the introduction (see lines 79 – 83), and carefully stated that all the calculations are for "perturbations from fresh NPF events upon the background aerosol that in itself may have been shaped by NPF earlier on, (lines 470 – 482 in the main text)". We also note that the growth of preexisting particles (that some may be from earlier NPF events) also contribute to the droplet number and present an analysis that deconvolves their effect on s_{max} and droplet number (see lines 520 – 531).

<u>Minor and technical issues:</u>

Please use .. paper.

Done.

lines 67-70: Compared … replaced with the newer one here.

Done.

line 73: Sipila et al. (2016, Nature, … consider adding that reference here.

We thank the reviewer for pointing this out. Done.

lines 91-92: Please mention explicitly that d_c refers to a critical diameter.

Done.

line 296: Cloud have both updrafts … in the paper.

Done.

lines 372-378: It took me … making the text a bit easier to follow.

This section has now been revised for clarity.

lines 380-383: This statement is incorrect. There ...size distribution measurements.

Thank you for pointing this out. We will replace the word "all" with "several" and the relevant references will be accordingly added.

line 396: "intermediate ions" is a commonly-used concept. What do the authors mean by "intermediate nucleation mode particles"?

The "intermediate nucleation mode particles" term used throughout the manuscript, corresponds to particles with diameters from 9 to 25 nm. Since we had no means to determine the intermediate negative-ion concentrations, this concept can be used as a quality test, in order to determine the initiation of a NPF event. The starting of the NPF is further confirmed by the evolution of the particle size distribution ("banana-shape pattern) when the new 9–nm particles appear and shift gradually towards larger sizes. We will make this point very clear.

lines 397-407: It is said that NPF starts at 8:30 and that these particles reach 100 nm at 21:30. This is not consistent with the given growth rate of 3.7 nm/h for nucleation mode particle. Does this mean that this particle population actually grows faster when reaching larger size, as mentioned in some other context later in the paper?

This is a good point. We will include the respective reference in order to avoid this confusing point. Thus, we are going to revise the text as: "Afterwards, particles continued to grow faster in size (Paasonen et al., 2018) for several hours, exceeding 100 nm in diameter at 21:30 LT.".

line 522: I suppose one of these velocities should be 0.6 m/s.

Indeed so! Thank you for pointing this out.

If Figures 3a and 3b are top of each other, it would be nice if their time axis matched with each other.

We will modify it in the revised version according to your suggestion.

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