The work by Kalkavouras et al. (2018) describes new metrics to evaluate the impact of new particle formation (NPF) on cloud condensation nuclei (CCN) budget and on cloud droplet number concentration (CDNC). The manuscript combines the analysis of an extended and valuable dataset, including both particle size distribution and chemical composition measured over 7 years at Finokalia, as well as model simulations to address the aspects related to CDNC. This manuscript aroused my interest and I believe it is worth publishing after some revisions. In its current form, there are several areas of the manuscript that need to be clarified, and in other areas the reader would benefit from additional information. Also, I believe that there are some inconsistencies between the different sections, and I think the authors contradict themselves in several areas of the manuscript. Finally, the distribution of information between the main text and the supplementary is sometimes questionable, and might be re-considered. In specific, CCN calculations performed at lower supersaturations (0.1%), which are expected to be more representative of real clouds, should be discussed in the main text. My detailed comments are listed below; they mainly concern the main text but the authors are encouraged to also take them into account to revise the abstract and the supplementary.

Comment 1: L55 & 58: It should be mentioned that Hyytiälä is located in Finland, in the boreal forest, and that the observatory of Chacaltaya is in Bolivia, at 5240 m a.s.l.

Comment 2: L70: Recent study by Kerminen et al. (2018) should be cited. Also, even if the present work does not aim at providing an exhaustive review of studies dedicated to marine environment, the papers by Sipilä et al. (2016) and Sellegri et al. (2016), which highlight the role of iodine in NPF, should be cited as well.

Comment 3: L92: The acronym d_c should be explicitly defined.

Comment 4: L109-113: For consistency, it should be mentioned which locations are investigated in the paper by Kerminen et al. (2012). Also, the sentence from L110 to 113 should be checked carefully, as it is confusing (I would suggest to remove the last part “and the maximum … during an event”).

Comment 5: L123: What do the “climate-relevant properties” refer to?

Comment 6: L129-133: The sentence should be rephrased.

Comment 7: L136-138: As suggested, this observation has already been reported, and should thus be supported by a reference. For instance, the paper by Leaitch et al. published in 1986 reported such observation.

Comment 8: L141-146: I would suggest to split the sentence into two parts, as it is too long in its current form. Also “depending” should be used instead of “depended”, and “reported” instead of “reporting”.

Comment 9: L146-149: It was thus already known/reported from the previous study by Kalkavouras et al. (2017) that discrepancies between CCN and CDNC enhancement arose partly from the supersaturations used for CCN calculations, which were too high compared to actual supersaturations observed in clouds. I thus wonder why, based on this result, the authors did not focus more on the CCN calculations performed at lower supersaturations (0.1%), which are discussed only in the supplementary.

Comment 10: L152-153: “continuous measurements of aerosol number size distributions and chemical composition”: based on the information provided in Section 2.2, the chemical composition of the particles was not measured between January 2011 and April 2012, is that correct? If so, the expression “continuous” should be removed from the introduction, before more information is provided on data availability in the next sections.

Comment 11: L155: “characterize the differences between nucleated particles”: what does that mean?

Comment 12: L158: “we consider all the issues”: I think this is too strong. As an example, if the sensitivity of CDNC calculation to updraft velocities is partly investigated with the use of two different values, the seasonality of such parameter and related effect on predicted CDNC is not discussed. I would thus recommend to use a more balanced expression, or at least to remove “all”.

Comment 13: L181-184: Please check the sentence; last part from “and thereafter…” is confusing.

Comment 14: L192-195: There is a word missing in the current form of the sentence: “sudden …?... of”. Also, I would suggest to clearly mention the particle growth process: “by a sudden increase of nucleation-mode particles concentration (…), and further growth of these freshly formed particles that lead to a continuous increase in larger…”

Comment 15: L197-200: From what I understand, the method reported here is not consistent with the equations 1-3. Indeed, based on these equations, the width and location of the three modes (nucleation, Aitken and accumulation) are kept constant (9-25 nm, 25-100 nm and 100-848 nm), and the particle concentration in each mode is calculated from the sum of the particle concentration in all the size bins of the corresponding diameter range. How does this relate to the use of a multi log-normal distribution function? Was this method used in a first approach to get the “average” diameter ranges which are used in this work? This needs to be clarified.

Comment 16: L205: I would suggest to slightly change the wording to “i_9, i_{100}, i_{848} refer to the SMPS size bins with mean (?) diameter 9, 100 and 848 nm, respectively”.

Comment 17: L216-218: The knowledge of the PM1 chemical composition is a key parameter in the present work. I would thus recommend to briefly recall the method from Bougiatoti et al. 2009. In specific, the limits/uncertainties associated to this method, and how they affect the calculation of CCN and CDNC should be discussed. Also, when estimating the organic fraction, which ratio of OM/OC was used?

Comment 18: L218-223: More information about ACSM measurements and data analysis should be provided:

- What type of ACSM was used (Quad/Tof)?
- Standard/capture vapourizer?
- Did you apply any collection efficiency correction?

Comment 19: L232: I would write “a top-bottom column temperature difference”, as if I am not mistaken (and even if it is quite straightforward!) the acronym T (and thus ΔT) has not been explained before in the text.

Comment 20: L236: What does “classified ammonium sulfate aerosol” mean?

Comment 21: L251: The equation should be given a number. Also I have several questions about the use of this equation:

- From what I understand, the main reason why to use this equation instead of a fixed d_c is because it takes into account the chemical composition of the particles *via* kappa. However, when only filter measurements were available, there was only one kappa value available per day, right? Wasn’t it so then that using the equation was in the end was very similar to using a constant d_c, as done in numerous previous studies?

- The variations of kappa appear to be quite limited on Fig. S3, so it is questionable how kappa actually affect d_c, and in the end, to which extent using the abovementioned equation improves the calculation of CCN concentration compared to the use of several fixed d_c. In other words, did you study, for a given supersaturation, the variations of d_c caused by the variations of kappa?
- In connection with comment 17, did you evaluate the uncertainty on kappa calculation arising from the use of PM10 chemical composition to derive information about PM1? Did you evaluate the "magnitude" of the uncertainty on CCN calculation related to the use of these indirect measurements (couple with the fact that the size dependence of kappa is not taken into account) compared to that of the older method, with "reasonable" fixed d_c?

I would at least suggest to clearly mention the uncertainty/limits of the method which are highlighted in the previous questions, and/or better emphasize the benefits that I may have missed!

Comment 22: L290-291: In connection to my previous comment: would it be possible, for each supersaturation, to get an average d_c from the CCN chamber measurements, then calculate the CCN concentrations corresponding to these "fixed" d_c (in a similar way as done in the previous studies) and finally evaluate the corresponding prediction error? This would, in my view, help to assess the benefit from introducing the kappa in the CCN calculation, as suggested in the present work, or at least give an idea of the "limits" of this approach.

Comment 23: L299-300: In connection with comment 9: "determine the cloud-relevant supersaturations for which CCN perturbation calculations are relevant". Such "relevant supersaturations" have already been reported by Kalkavouras et al. (2017), so, again, calculations performed at 0.1% should in my view be the main focus of this work, and should be used to further link/compare CCN and CDNC results obtained in sections 3.2 and 3.3.

Also, did the author get the chance to evaluate the relevance of the predicted N_d against for instance airborne in-situ measurements conducted in the vicinity of Finokalia?

Comment 24: L316: "vigorous boundary layer": do the author mean "turbulent"?

Comment 25: L332-336: The sentence should be checked and rephrased; also "were used" (L334) should be changed to "was used".

Comment 26: L344-354: Few suggestions:

- I would recall the periods during which each measurement technique was used;
- Wouldn't it be possible to summarize all the values on a plot, using pie charts for instance?
- It is not clear to me which instrument was used to derive the seasonal values discussed from L351 to 354;
- I was surprised to read that highest organic contribution was observed during wintertime; I would be curious to learn about the main sources during this time of the year.

Comment 27: L358-359: I do not understand this sentence: in my view the absolute concentrations should not affect kappa, only the fractions (i.e. "epsilon") should matter.

Comment 28: L364-368: The decrease of kappa between 6:00 and 9:00 LT is not obvious for me on Fig. S3… Also, would it be possible to add to Fig. S3 the time series of sulfate and organics measured with the ACSM, to support the hypotheses regarding the variations of kappa?

Comment 29: L370: Is the kappa difference of 0.2 kappa units calculated from average values? Because from Fig. S3, it seems that the difference can reach almost 0.4 (during the night and at the end of the afternoon).

Comment 30: L373-374: In connection with comment 28, the convergence which is reported in this sentence is also not obvious for me.

Comment 31: L374-378: This sentence is confusing me a lot, as, in my view, it conflicts with some ideas which are discussed elsewhere in the paper:

- "This constant character of the chemically derived kappa may be an evidence that using prescribed levels of supersaturation or critical diameters to calculate CCN concentrations can provide a biased influence of NPF events on CCN". In my view, the observation of the constant character of kappa does not indicate at this stage that the CCN predictions obtained from prescribed levels of supersaturation or prescribed diameters are biased; it only highlights the fact that both approaches are finally very similar, since the limited variations of kappa lead in the end to almost prescribed d_c. This assertion is even more surprising that Fig. S2 and L281-294 highlight a pretty good agreement between CCN prediction from ACSM/SMPS data and direct CCN measurements.
- "since there is a clear dependence between the chemical composition and the size of a particle": isn't is conflicting with L275-276 ("a size dependant consideration of hygroscopicity is therefore deemed unnecessary") and end of Section 2.4?

Comment 32: L386-389: The phrasing of the sentence is quite confusing; also, I wonder if it is relevant to apply this classification, which is exclusively based on measurements conducted in Pittsburgh, to measurements conducted at Finokalia, where particle concentrations and NPF event characteristics are most likely different. I would remove the sentence because I don't think it provides valuable information.

Comment 33: L396: What does "intermediate nucleation mode particles" mean?

Comment 34: L411: "the time series of the aerosol size distribution and chemical composition": since different datasets/instruments/measurement techniques were involved in this work, I would clearly recall that when filter measurements were used, there was only one kappa value available per day, to keep the message as clear as possible.

Comment 35: L415: I think there is a space missing: "supersaturations" -> "supersaturation *s*"

Comment 36: L417: Information in the brackets is not useful.

Comment 37: L420: It might be useful to also indicate t_{start} on Fig. 3a. Also, the expression "Prior to 8:30 LT and 5 hours later" is not clear to me.

Comment 38: L429: "dividing" instead of "diving"?

Comment 39: L431: Over which period was the average value calculated? Full day?

Comment 40: L436-437: "from the influence of NPF on the larger supersaturations": even if I get the message, I think it would be more correct to change the wording to something like "from the influence of NPF on the production of particles which activate at larger supersaturations".

Comment 41: L438: On the 29th of August, the influence of NPF on CCN production is said to terminate at 21:30; was it decided that this "end time" would systematically be identified on the day of the NPF

event, or did the authors extended their research period to the next day, to document growth processes possibly spanning on several days?

Comment 42: L445-446: "this variation of R_s can be equivalent to the percentage contribution of CCN owing to NPF": I would suggest to change to wording to something like "This variation of Rs indicates, for each supersaturation value, the increase of the CCN concentration related/due to particles originating from NPF".

Comment 43: L453: "the time which the new particles after the t_{start} are able to grow": I would suggest to rephrase this part of the sentence to make it clearer.

Comment 44: L454-455: "This time fluctuates from 2.7 to 10.5 h in the 1.0-0.38%". The value of 2.7 h was obtained assuming an initial diameter of 25 nm for the newly formed particles at t_{start}, is that right? If so, I am not convinced by this approach, since I would expect most of the particles in the nucleation mode to be smaller than this upper limit at t_{start}.

Also, how did the authors get the upper value of 10.5 h? For me the longest time delay should correspond to particles with a diameter of 9 nm at t_{start}, which then need to reach 67 nm to be able to act as CCN at s = 0.38%, i.e. + 58 nm. Considering a GR of 3.7 nm/h, I find that it takes approximatively 7 hours for the particles to reach this d_c. Repeating the same calculation with initial diameter of 25 nm leads to a bit more than 11 hours. The hypotheses used for this calculation should be clarified.

Comment 45: L456: "start to feel the influence of NPF". In connection to comment 40, I also get the message but I would rephrase this part of the sentence, and refer more to the time it takes for the newly formed particles to grow to d_c = 67 nm.

Comment 46: L456-458: "t_{dec} is later for supersaturations below 0.7%": I do not understand the meaning of this sentence, since based on the definition from L425-429, there is one single value of t_{dec} per event, which is derived from all supersaturations. Do the authors mean that it takes longer time to observe the influence of NPF on the concentration of particles able to act as CCN at lower supersaturations, as it is expected that those need to grow to larger sizes? Also, the link with the second part of the sentence is not clear to me.

Comment 47: L461-462: "indicating that the newly formed particles in this size range may exhibit similar chemical composition". Similar chemical composition at all sizes was assumed from the beginning of the calculation with the use of a single kappa for all sizes, wasn't it?! (L275).

Comment 48: L464-466: How did the authors get the reported values? By averaging all R_s between 13:30 and 21:30? If so, in connection to comment 42, I would again talk more about an increase of the CCN concentration due to NPF rather than a contribution of NPF, and I would suggest to further check this aspect throughout the manuscript.

Comment 49: L466-467: "since the d_c … respectively": I do not think this is a proper explanation, I would rather say "consistent with similar R_c observed in the same size range, as mentioned above".

Comment 50: L469-476: I am not convinced by the suggested correction process for the two main reasons which are developed below:

- Background particles possibly contributing to CCN population together with growing particles originating from NPF are those which were already large enough before t_{start}/t_{dec}, not those in the nucleation mode before t_{start}. And, by the way, particle concentration in the nucleation mode should be around zero before t_{start}, as by definition those particles originate from nucleation.

- Also, if I am not mistaken, this paper discusses the CCN concentration increase from a reference concentration taken at t_{start}, which, I expect, already includes some contribution of background particles. I would thus say there is no need to apply any correction. The only bias, which is complex to evaluate but should still be mentioned, is caused by the possible appearance of large particles not originating from NPF between t_{start} and end of NPF influence on CCN concentration (21:30 in this case), as those can impact the variations of the CCN concentration predicted at a certain (most likely low) supersaturation during this time period.

Comment 51: L478: I would suggest to remove "to the R_s and subsequently".

Comment 52: L488: "for" should be removed.

Comment 53: L486-491: I would have expected particle GR to be the main factor determining the time delay between t_{start} and t_{dec}, but the seasonal variation of this time delay (similar in winter, spring and summer, and lower compared to autumn) is not consistent with that of the GR reported by Kalivitis et al. (2018) (higher GR in summer, lower in winter and spring). Could the author comment on this aspect?

Comment 54: L494-495: "at cloud supersaturations encountered in this environment". To me, this sentence conflicts with what I think is a main message of the authors, yet already reported by Kalkavouras et al. (2017): L563, "the actual cloud supersaturation being much lower than the prescribed levels in the CCN analysis". (Also L33)

Comment 55: L496: Should be 3.3 instead of 3.4.

Comment 56: L504: I think the time interval between 8:30-11:00 is not correct to refer to the "growth hours" of the episode. This is at least not consistent with the fact that the influence of growing newly formed particles on CCN population is seen between 13:30 and 21:30.

Comment 57: L508: Space missing between "increases" and "4.7".

Comment 58: L509-515: "Both trends are related to decreases in accumulation mode aerosol number": the decrease of accumulation mode particle concentration is not clearly visible on Fig. 1. Also, I wonder why at this stage of the analysis the variations of N_d are related to accumulation mode particles only, since the results of the previous section suggest a major contribution of Aitken mode particles to CCN population (d_c < 67 nm). To support their assumption, the authors would first need to discuss the inconsistency between the supersaturations used to predict CCN concentrations and s_{max} retrieved by the model; s_{max} being lower, it implies that particles need to grow to larger sizes to effectively act as CCN, and yes, in the end they most likely belong to accumulation instead of Aitken mode.

« as the latter has not had the chance to influence particles that act as CCN in clouds": again, I think this is not correct at this stage of the study, since the previous section highlights the influence of NPF on CCN concentration already from t_{dec}, i.e. 13:30. The supersaturation inconsistencies recalled above are also needed to further explain/clarify this aspect. Particles can act at CCN already from t_{dec}, but in the presence of supersaturations which are most likely significantly higher than those predicted by the model.

Considering the different supersaturations discussed in sections 3.2 and 3.3, I think it is finally complex to establish/comment on the link between t_{dec} and t_{nd}.

Another point: how do the authors determine the beginning of NPF influence on N_d at 17:25? From Fig.5, it seems that the most significant increase of N_d is seen from ~21:00. Now looking at Fig. 1.A, this time coincides with the time at which the NPF event is somewhat interrupted, suggesting that the

particles contributing to the increase of N_d could finally not be related to NPF. Could the authors comment on that?

Comment 59: L519-529: It is complex for me to understand this part of the analysis. Specific comment on L527, "Since N_d does not increase significantly until midnight": looking at Fig. 5 I would say it does, at least until 23:00. Second part of the sentence is also not clear to me.

Comment 60: L533: I do not understand how the authors calculate the value of 30 cm^{-3}.

Comment 61: L534: In connection to comment 41, why is the analysis stopped at midnight? Again, if the analysis is limited to the day of the event, this has to be mentioned, also the reason why.

Comment 62: L535-538: I think the length of the dataset is a strong point of this work, so I would really suggest to discuss more the "statistics" in the main text. In general, the supplementary includes valuable information, from which the reader would benefit more if it was partly moved to the main text. This comment also applies to CCN related calculations reported in the previous section (see for instance comments 9 and 23).

Comment 63: L548: "accurately" is in my view too strong.

Comment 64: L550, 553: time delays between t_{start} and t_{dec} are different from those reported in Sect. 3.2. Also, it would be better to have a uniform notation to report durations (L550, 553 and 554).

Comment 65: L558-559: "the impact of NPF on N_d differs considerably from the CCN based analysis". I do not completely agree with this sentence, as it compares two different variables (CCN concentration and N_d) calculated using different hypotheses.

Comment 66: L559: "Regardless of season". This assessment is a bit too strong in my view, as a possible seasonal variation of some parameters such as vertical velocity (and thus further effect on N_d calculation) has not been discussed. Also, I would not talk about "typical boundary layer clouds"; L310 indicates "in cloudy boundary layer in the region".

Comment 67: L569-570: Please refer to comment 50.

Comment 68: L571-575: I would not say this is a striking consequence; I would rather say it is expected, particles need to grow to large-enough sizes to be activated into droplets, which takes time, and this is even more the case when supersaturations get lower. I am also not sure about the last part of the first sentence "N_d is insensitive to increase in CCN during the course of an event… vapor": again, during the course of the event, CCN population possibly activating at higher supersaturation is increased first, and as particles are getting bigger towards the end of the event, they can activate at lower supersaturations.

Comment 69: L579-580: Wasn't it already a result from Kalkavouras et al. (2017)? See also comments 9 and 23.

Comment 70: L581: The influence of what?

Comment 71: L584: "highly effective paradigm" is too strong for me; if new metrics are introduced for quantifying the temporal aspect of NPF influence on CCN production, CCN calculation itself relies on the use of prescribed supersaturation, which is simultaneously reported to be hazardous (eg L525, 581).

Comment 72: L741-749: Check alphabetical order.

Comment 73: Fig. 1: Caption, (b), I would remove “differences”, and simply refer to the time series of the particle concentration at different time resolutions.

Comment 74: Fig. 3: it would help to have similar scale on the x axis of Fig. 3 a and b.

Comment 75: Fig. S1: x axis label: “ACSM” instead of “ASCM”. Also, adding the 1:1 line would help to interpret the results.

Comment 76: Fig. S3: Caption: space missing between “the” and “kappa”.

Comment 77: Table S2: I would suggest to clearly explain what “bef” and “aft” refer to, as these are used in several tables of the supplementary. Also, in the caption, it is not clear to which “relative contribution” the authors are referring to. I would clearly mention it is the increase of Rs (%) observed after t_{dec}, and clearly indicate it corresponds to the “Change” column.

Comment 78: Table S5: If I am not mistaken, the authors never refer to Table S5, neither in the main text nor in the supplementary. This table report values which would have been very interesting to discuss more in the main text, in specific those calculated for s = 0.1%, as this supersaturation is thought to be more “representative” of real clouds. Also, more information would be needed regarding the calculation of the N_{CCN} values, as I think it has not been explained elsewhere: over which time period (average between t_{dec} and end of NPF influence?)? Any correction applied?

Comment 79: Supplementary: several spaces missing in different places.

Bibliography

Bougiatioti, A., Fountoukis, C., Kalivitis, N., Pandis, S. N., Nenes, A., and Mihalopoulos, N.: Cloud condensation nuclei measurements in the marine boundary layer of the Eastern Mediterranean: CCN closure and droplet growth kinetics, Atmos. Chem. Phys., 9, 7053-7066, https://doi.org/10.5194/acp-9-7053-2009, 2009.

Kalivitis, N., Kerminen, V.-M., Kouvarakis, G., Stavroulas, I., Tzitzikalaki, E., Kalkavouras, P., Daskalakis, N., Myriokefalitakis, S., Bougiatioti, A., Manninen, H. E., Roldin, P., Petäjä, T., Boy, M., Kulmala, M., Kanakidou, M., and Mihalopoulos, N.: Formation and growth of atmospheric nanoparticles in the eastern Mediterranean: Results from long-term measurements and process simulations, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-229, in review, 2018.

Kalkavouras, P., Bossioli, E., Bezantakos, S., Bougiatioti, A., Kalivitis, N., Stavroulas, I., Kouvarakis, G., Protonotariou, A. P., Dandou, A., Biskos, G., Mihalopoulos, N., Nenes, A., and Tombrou, M.: New particle formation in the southern Aegean Sea during the Etesians: importance for CCN production and cloud droplet number, Atmos. Chem. Phys., 17, 175-192, https://doi.org/10.5194/acp-17-175-2017, 2017.

Kerminen, V.-M., Paramonov, M., Anttila, T., Riipinen, I., Fountoukis, C., Korhonen, H., Asmi, E., Laakso, L., Lihavainen, H., Swietlicki, E., Svenningsson, B., Asmi, A., Pandis, S. N., Kulmala, M., and Petäjä, T.: Cloud condensation nuclei production associated with atmospheric nucleation: a synthesis based on existing literature and new results, Atmos. Chem. Phys., 12, 12037-12059, https://doi.org/10.5194/acp-12-12037-2012, 2012.

Kerminen, V.-M., Chen, X., Vakkari, V., Petäjä, T., Kulmala, M., and Bianchi, F.: Atmospheric new particle formation and growth: review of field observations, Environ. Res. Lett., 10, https://doi.org/10.1088/1748-9326/aadf3c, 2018.

K. Sellegri, J. Pey, C. Rose, A. Culot, H.L. DeWitt, S. Mas, A.N. Schwier, B. Temime-Roussel, B. Charriere, A. Saiz-Lopez, A. S. Mahajan, D. Parin, A. Kukui, R. Sempere, B. D'anna, and N. Marchand, Evidence of atmospheric nanoparticle formation from emissions of marine microorganisms, Geophys. Res. Lett., 43(12), 2016GL069389, doi:10.1002/2016GL069389, 2016.

W. R. Leaitch, J. W. Strapp and *G.* A. Isaac: Cloud droplet nucleation and cloud scavenging of aerosol sulphate in polluted atmospheres, Tellus, *388,* 328-344, 1986.

Mikko Sipilä, Nina Sarnela, Tuija Jokinen, Henning Henschel, Heikki Junninen, Jenni Kontkanen, Stefanie Richters, Juha Kangasluoma, Alessandro Franchin, Otso Peräkylä, Matti P. Rissanen, Mikael Ehn, Hanna Vehkamäki, Theo Kurten, Torsten Berndt, Tuukka Petäjä, Douglas Worsnop, Darius Ceburnis, Veli-Matti Kerminen, Markku Kulmala, and Colin O'Dowd : Molecular scale evidence of new particle formation *via* sequential addition of HIO3, Nature, 537(7621): 532–534, doi:10.1038/nature19314, 2016.