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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Regional New Particle Formation as Modulators of Cloud Condensation Nuclei and Cloud Droplet Number in the Eastern Mediterranean" *by* Panayiotis Kalkavouras et al.

Anonymous Referee #1

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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.

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.

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.

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.

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.

Minor and technical issues

Please use the term "cloud droplet number concentration" instead of "cloud droplet number" throughout the paper.

lines 67-70: Compared with Kulmala and Kerminen (2008), the topics of these lines are covered in much more detail the recent review by Kerminen et al (2018, Environ. Res. Lett. 13, 103003, https://doi.org/10.1088/1748-9326/aadf3c). The older review could be replaced with the newer one here.

line 73: Sipila et al. (2016, Nature, 537, 532-534, doi:10.1038/nature19314) provide the most comprehensive mechanistic description of coastal NPF presented so far. The authors might consider adding that reference here.

lines 91-92: please mention explicitly that dc refers to a critical diameter.

line 296: cloud have both updrafts and downdrafts, so I am sure that "cloud vertical velocity" is the proper wording here. The authors should maybe stick with "cloud updraft velocity" here as done elsewhere in the paper.

lines 372-378: It took me a while to understand the message of this discussion. The main point appears to be that hygroscopic properties (kappa) of a particle population tends to depend on the particle size, but this feature is not revealed by bulk aerosol composition measurements like those done with ACSM. And that this has consequences in interpreting the data. Please consider making the text a bit easier to follow.

lines 380-383: This statement is incorrect. There are at least 2 long-term studies in which the contribution of NPF to CCN has been investigated using direct CCN measurements (Sihto et al. 2011 already cited in this paper, Dameto de Espana et al.

2017, Atmos. Environ. 164, 289-298), not just particle number size distribution measurements.

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

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 the this particle population actually grows faster when reaching larger size, as mentioned in some other context later in the paper?

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

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

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