

Interactive comment on “New particle growth and shrinkage observed in subtropical environments” by L.-H. Young et al.

Anonymous Referee #2

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This manuscript analyses interesting, and quite rarely reported, observations of aerosol shrinkage following their prior formation and growth. The paper is worth to be published, but needs to be partly re-written to highlight this observation and to avoid over-interpreting some other aspects related to atmospheric aerosol formation. My detailed comments are given below.

Response:

We are grateful to Anonymous Referee #2 for providing insightful comments that helped us improve our manuscript substantially. Our point-to-point responses to the reviewer’s comments are provided and highlighted in blue-colored font, as follows.

Major issues:

14 events were reported altogether at 4 sites, and the events were further divided into type A and B events. Because of the very low number of certain type of events at any single site, the authors should avoid making too general conclusions about the results. The number of cases is simply too low. The main emphasis of the paper should be on the particle shrinkage and possible factors causing it.

Response:

Because of the poor statistics, as suggested by the reviewer, the classification of a small number of NPF events into subsets (Types A and B) has been removed. In effect, we have changed our focus to the differences between non-event and event days, as well as the particle shrinkage events.

The analysis of the formation rates of 1 nm particles (section 3.2) should be taken away from the paper for two reasons: i) there is a very large uncertainty in these values as there is no experimental information on particles concentrations or growth rates between 1 and 10 nm, ii) the number of reported events is very low for any meaningful comparison to other sites.

Response:

We have revised the manuscript to focus on the J_{10} , whereas the modeled J_1 are retained in the manuscript as supplementary information. In addition, we have included Korhonen et al. (2011)’s work to caution the readers about the potential uncertainties in deriving the J_1 because of the lack of knowledge on the size dependence of particle growth rate below 10 nm as well as the possible erroneous assumptions about the initial cluster size of ~1 nm. Specifically, the decreased growth rate with decreasing particle size and the assumption of a possible too small critical

cluster size of 1 nm together likely leads to an overestimation of mean nucleation rates by the PARGAN inversion method. Nevertheless, unless equipped with instruments capable of detecting ~1 nm particles, we suggest that the highly time-resolved PARGAN approach still provides useful information about the intensity of atmospheric nucleation that can be used as a reference and benchmark for future improvement studies.

Cited reference:

Korhonen, H., Sihto, S. L., Kerminen, V. M. and Lehtinen, K. E. J.: Evaluation of the accuracy of analysis tools for atmospheric new particle formation, *Atmos. Chem. Phys.*, 11(7), 3051–3066, 2011.

In principle, I like the case study-approach chosen here. However, having 7 figures (figure 3-9) with exactly the same and very detailed information is definitely too much. Two carefully-selected full figures like that should be enough. For the rest of the cases, it is sufficient to plot the first panel of the figure, from which the time evolution of the particle number can be easily seen.

Response:

We agree with the reviewer's suggestions on the careful selection of case studies. As a result, we have revised the manuscript to include two growth events (with each representing the morning and midday NPF, respectively) and two shrinkage events (with each representing the positive and negative correlation, respectively, between the number concentration of 10-25 nm particles, N_{10-25} , and the particle mode diameter, D_{mode}). Although the underlying mechanisms were not clearly elucidated in the present study, we suggest that the timing at which the NPF commence is an important factor because it provides clues as to what conditions (e.g., atmospheric mixing or photochemistry) are conducive to NPF at the study sites. Similarly, the increasing (or decreasing) N_{10-25} with increasing D_{mode} provides possible mechanisms (e.g., evaporation) related/leading to particle shrinkage.

Minor issues:

The statement particles larger than 50 nm act as CCN should be backed up with a reference.

Response:

The statement of particles larger than 50 nm act as CCN is based on the study by Dusek et al. (2006). They reported that particles < 40 nm require unrealistic high saturation for activation, whereas particles > 120 nm generally activated at all studied values of saturation regardless of composition. As a result, they referred to particles in the size range of 50-150 nm as CCN-relevant size range.

Cited reference:

Dusek, U., Frank, G., Hildebrandt, L., Curtius, J., Schneider, J., Walter, S., Chand, D., Drewnick, F., Hings, S. and Jung, D.: Size matters more than chemistry for cloud-nucleating ability of aerosol particles, *Science*, 312(5778), 1375, 2006.

The total event frequency appears low. Was this because only the most intense events were analyzed, or was the event frequency low also when considering the traditional ways of classifying the event in the literature?

Response:

The NPF frequency is low mainly because of the following two reasons. First, only the most intense regional events (i.e., NPF followed by growth over 1.5 hr) were analyzed, as described on p.18613, lines 1-7, which is in accordance to the classification scheme proposed by Dal Maso et al. (2005). This ensures that the particle formation and growth rates can be determined with good a confidence level. Second, the study periods were relatively short and thus the NPF frequency may not be representative to that actual NPF frequency. Kulmala et al. (2012) recommended that a minimum measurement period of several weeks is needed for individual event studies, and one year for determining the seasonal NPF frequency. Accordingly, the present study falls into the former class of NPF case studies.

Cited references:

Dal Maso, M., Kulmala, M., Riipinen, I., Wagner, R., Hussein, T., Aalto, P. P. and Lehtinen, K. E. J.: Formation and growth of fresh atmospheric aerosols: eight years of aerosol size distribution data from SMEAR II, Hyytiälä, Finland, *Boreal Environ. Res.*, 10(5), 323–336, 2005.

Kulmala, M., Petäjä, T., Nieminen, T., Sipilä, M., Manninen, H. E., Lehtipalo, K., Dal Maso, M., Aalto, P. P., Junninen, H., Paasonen, P., Riipinen, I., et al.: Measurement of the nucleation of atmospheric aerosol particles, *Nat Protoc*, 7(9), 1651–1667, 2012.