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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Frequency of new particle formation events in the urban Mediterranean climate" *by* M. Brines et al.

Anonymous Referee #1

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The MS mainly deals with the evaluation of SMPS data sets for 5 measurement sites by k-means clustering analysis. The idea of the study is good, its goals are relevant, timely and of interest for the international scientific community in the field. Unfortunately, the work has not been performed on a scientific level that merits the high standard of the ACP, and it contains several confusions and mistakes.

1. Selection of the measurement sites and corresponding time intervals is not justified. Brisbane does not belong to the Mediterranean climate zone as shown in Fig. 1, which is in contradiction with the title. (The source of Fig. 1 is not given and the type of the climate classification system is not mentioned.) Los Angeles can not be accepted as well since a 3-month long data set is only available for it, which A) does not cover one full year, and therefore the seasonality in the nucleation frequency (which is obvious

in many areas) is disregarded, and B) it is also much shorter than for the other sites, thus 1 or 2 years. Furthermore, the measurements in Rome were performed at a regional background site (p. 6), which is again in contrast with the title, and questions the representative character of the conclusions for urban areas.

2. It is generally and well accepted that frequency of the new particle formation event is determined on a daily basis, and that it represents the number of nucleation days with respect to all/relevant days on a certain time scale (week, month or year). The title of the paper is misleading not only because the concept of the frequency is completely different here, but - more importantly - since it can not be related to the nucleation event itself (see also comment 3). Instead, it expresses the time share of the particle growth process. At the same time, it is the end of the growth process that is difficult to determine in urban environments due to, for instance, substantial emissions, and therefore, the frequency concept suggested here is doubted.

3. The lower diameter measurement limits (between 10.2 and 17. 5 nm) and the corresponding measurement diameter interval make the evaluation of the atmospheric nucleation events rather difficult in particular in cities since the most valuable diameter range, namely the interval below 10 nm is completely missing. As a consequence, the authors show a contour plot in Fig. 5 for Rome as a nucleation event although there is no indication of the nucleation mode (below 10-20 nm), and the elevated concentrations only appear above 20 nm, which is typical for emissions. This all questions if there was atmospheric nucleation at all that day. Such an unusual atmospheric event can not be classified or regarded as nucleation without firm and detailed explanations and evidence. Thus, the conclusions draw at a later stage are also not plausible. Let me also mention here that the heading of Fig. 5 "Daily average SMPS size distributions on a nucleation day . . ." seems to be obscure similar to many other formulations (p. 2: collected size distributions, p. 6: data were sampled, title of section 2.2.1, etc.) in the text, which may indicate that the MS was not elaborated carefully and by all co-authors.

4. It is not described at all how the number of representative clusters between 7 and

15 was reduced “after a careful consideration” (p. 8) to 4-7, which could be a critical issue, and lacks objectivity in its present form.

5. It is unusual to use “traffic-related nucleation mode” (e.g. on p. 9) because the particles which are formed within the source, plume or exhaust are considered as primary particles contained in the Aitken mode in contrast to the nucleated particles contained in the nucleation mode. The present reviewer admits that this can be somewhat more complex (see Robinson et al., Science 315, 1259-1262, 2007) but the usage of such expression without further specific explanations is not tolerable.

6. Fig. 2 shows particle number size distributions that resulted from k-means clustering. After a detailed examination of many curves, the readers can wonder if resolving the distributions of atmospheric aerosol particles into two modes is indeed realistic, or in other words, whether the clusters T1, T2 and T3 containing 2 modes each at 1) 20-40 nm and 70-130 nm, 2) 20-40 nm and 60-90 nm, and 3) 10-20 nm and 50-80 nm are indeed different.

7. In relation to comment 6, a sensitivity analysis or arguments should have been added on the uncertainty of some results. Without these, it can be questioned whether the frequencies of 6% (section 3.1.3) or 7% (section 3.1.4) are significant or just within the uncertainly limits.

Several comments listed above represent excluding criteria or arguments for rejection, and it is thought that the MS needs such an extensive improvement which can only be realised within the frame of a new submission.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 26463, 2014.