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Comment on acp-2022-126

Anonymous Referee #2

Referee comment on "Annual Cycle of Hygroscopic Properties and Mixing State of the Suburban Aerosol in Athens, Greece" by Christina Spitieri et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-126-RC2, 2022

Recommendation

The paper presents very valuable and rare 1-year data of hygroscopic growth factors of aerosol particles as function of size in a new environment, namely an urban background area in southern Europe. I recommend to accept the paper upon major revisions.

Summary of recommendations for major revision:

The paper misses GF calculations for 100-150 nm diameter. Please give reason for this. I acknowledge the difficulty in these kinds of measurements, but I require an honest explanation of the missing size range, which is very important for cloud condensation nuclei. Otherwise, the reader might think there was a scientific reason to leave these measurements out.

The analysis of GF is much too long and should be substantially shortened in number of figures and in the analysis, which requires a substantial new layout of the written text. The paper should be shortened by at least 1/3 of its current size in the number of words, and at least a half of the figures and tables should be removed in the paper and supplementary information. The readability is very hard at the current state and contains repeatability of similar messages (although shown with new types of results and analysis approach).

Some of the analysis of the results contains rather speculative discussions on the reason for high or low GFs. A more detailed analysis with trajectory data, wind speed data, and other meteorological data and a detailed look on individual days with particle number size distribution data is needed to reach firmer conclusions. However, without compromising

the obligatory shortening of the paper.

Detailed revision and comments

Length of paper:

Several of the figures could be removed (as a very good example Figure 10), which also goes for the figures in the supplementary information. For the analysis, one could for example mention the GF for the first time for all diameters at the same time. Then, one could focus on each individual dry particle diameter and summarize the findings around this particle size, and not mention all the different parameters and circumstances around it (GF, GF PDFs, sigma-values, diurnal variation, seasonal variation, less, intermediate, more hygroscopic modes, relative number fraction of the different hygroscopic modes, meteorological influence, and so on) if it doesn't give new substantial information. Alternatively, the authors can choose another strategy as well for the shortening of the text and removal of figures.

Due to the length of all information, it is very difficult for me as reviewer to make a decision on which figures and analyses to shorten. The authors are more familiar with their own data sets and results, and hence I leave it to the authors to make this prioritization.

Analysis of GFs:

The reason for some of the interpretation of GFs is sometimes speculative. How is it even theoretically possible that the 30 nm diameter particles are more hygroscopic than the 250 nm particles if they come from traffic exhaust? Normally one would expect very high number fraction of hydrophobic particles from relatively fresh fossil fuel combustion at around 30 nm in an urban area (e.g. Guo et al., 2020, https://www.pnas.org/doi/full/10.1073/pnas.1916366117; Titta et al., 2010, doi:10.1016/j.atmosenv.2009.06.021, Kristensson et al., 2013, https://aaqr.org/articles/aaqr-12-07-oa-0194 and many others). You have provided some context to this, explaining that some of the 30 nm hygroscopic particles might come from new particle formation events and that the traffic exhaust particles are aged. But you have to provide more detailed analysis to be able to come to this conclusion: Trajectory analysis (trajectories can be downloaded for free from the Hysplit site) if the air really comes from Athens and under what weather conditions and how long time it took for the air to arrive to the site from Athens and the phochemistry activity with meteorological parameters (for ageing purposes), and closer look at individual size distributions on individual days to see if it resembles a traffic exhaust particle size distribution, or new particle formation or something else. It is not enough to look at the average size distribution of clusters like in Figure 11, since the averaging of several size distributions might mask the shape of the individual size distributions. Besides, a look on an individual day would reveal if it is a new particle formation event day or not.

A closer look on all of the clusters in Figure 11 for individual days is also necessary to make correct conclusions. To me it seems that the interpretation of the sources of different clusters and their typical sizes is not correct or highly speculative. Based on the size distribution shape, the diurnal variation and the wind direction doesn't lead to the conclusions about the origin of the clusters. For example, the second cluster is not at all nocturnal, and even seems to be more of a traffic exhaust related cluster than cluster 1 due to the association with morning and evening hours, which could be representative of morning and evening traffic. Maybe, the clustering does not even give a valid representation of different representative aerosol types. Maybe you should consider to abandon this analysis and make a manual analysis instead of how air masses influence the size distributions and in turn the GFs as suggested previously?

Another example of inconclusive interpretation is the sub-10 nm diameter particles log-normal modes that you present in Figure 11. You have to take a closer look on the possible sources of this mode: Is it particles from traffic exhaust that have nucleated some time after the emissions? It is probably not primary emitted traffic particles in Athens, because the maximum for such a mode should be significantly higher than 20 nm diameter at the time it reaches the site after 1 hour of ageing or similar. Again, a closer look at size distributions in connection with trajectories could reveal the reason for their appearance. Please also explain cluster 5 in a clearer way, it seems to contain contradictory information about the sub-10 nm diameter mode when speaking about cooling of exhaust gas.

Grammar:

Some grammatical improvements can be made, for example: "As a direct effect, aerosol particles interact with solar radiation through light absorption and scattering, inducing a positive or negative radiation forcing". "As a direct effect" sounds strange. Another example: "The hygroscopic properties of atmospheric particles are strongly related to particle chemical composition (Gunthe et al., 2009; Gysel et al., 2007), while they undergo continuous changes over particle lifetime". Why do you write "while" in this sentence? These sentences sound a bit strange, and such are found throughout the paper. This needs to be corrected.

Comprehension:

Chapter 2.3.3 is hard to understand. I know what you mean, since I have been doing similar things. But, not sure that people who haven't done this before will understand your method approach. Please describe it in a few more sentences to make it clearer.