Reply to reviewer #2

We thank the reviewer for the insightful comments and for considering the manuscript worth publishing. Below, find our point-to-point response to specific comments.

Detailed response to comments (blue font)**:**

Summary of recommendations for major revision:

Please note that we have isolated all comments in this summary and provided a response as follows:

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.

We acknowledge that this is a valid question one may raise in this context. However, as the reviewer indicates there are practical difficulties when implementing HTDMA measurements. One of this is the limitations arising from selecting several representative size fractions to study but at the same time obtaining data in a relatively high time resolution. Having targeted the hourly time interval and the need to have at least three five minute size distributions (this is the length of an SMPS distribution scan) we could have 4 selected sizes (30nm, 50nm, 80nm, 250nm). This selection may appear ad hoc but it reflects are previous experience with the moments of the number size distribution as observed previously (Vratolis et al., 2019). It is confirmed in the cluster analysis appearing in Figure 11, that we cannot detect an individual mode of particles indicating different physical properties in the range in question (100-150 nm). Please observe in this figure that all particles in this range are included mostly in the separate accumulation mode (100-550 nm). And in any case, 80 nm is in the logarithmic scale is very close to 100 nm. We expect that it is now understandable how the selection of these size was made.

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).

Following up on these suggestions we have removed Figure 5 and Figure 10 and we have improved the readability of the document by shortening some discussion. However, the reviewer should consider the suggestions by the first reviewer where additional information is required and further clarification of the figures, while extensive figures are suggested to be included. In order to satisfy both suggestions we have moved a considerable part of information in the supplement and expect that the manuscript is now very much improved.

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.

Please consider that the explanations provided on the levels of growth factors are based on the literature from previous studies which have been conducted at DEM station and on the specific characteristics of the microclimate of the city of Athens. Athens is a very large metropolitan area (approx. 5 million people) located in a basin where the diurnal pattern of transport and mixing of particle within the basin is responsible for the most of the particles we observe at DEM station in Athens. Considering that no major city or pollution source is located as far as at least 100 km we do not except that the fine short-lived aerosol number fractions (with the exception of the accumulation mode) can be characterized by back trajectory analysis. Large events like fires and desert dust transport events are the topic of this study. Valuable information can be derived by local meteorological wind speed and wind direction data. Therefore, a more detailed analysis of meteorological data will be performed for a more detailed explanation about the levels of growth factor values and included in the supplement.

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.

Following up this suggestion we have removed figure 5 and figure 10. However, reviewer 1 have requested many additional information to be included especially in the supplement. The supplement does not affect the length of the paper and we have not received any indication by the editor or the journal production that this length is problematic. The second suggestion to remove the different parameters calculated from the inversion of HTDMA data are an essential part of the paper, where this type of analysis allows us to resolve for the first time in a suburban area the state of mixing of the fine aerosol size fraction.

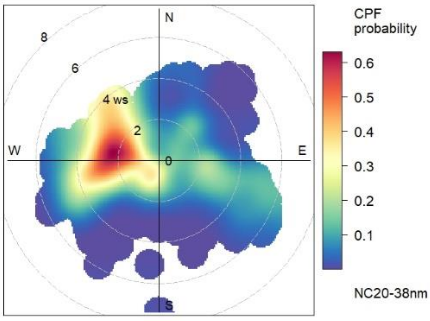
Analysis of GFs:

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 photochemistry 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.

We would like to demonstrate why our findings and interpretation is not speculative and clarify these points as well as add additional data and discussion in the manuscript so that we respond to the points raised by the reviewer.

Theoretical calculation for hygroscopicity and activation of particles is not straightforward for complex aerosol populations whereas it in our case there is variable state of mixing (different chemical composition) regarding aerosol hygroscopicity even for the same particle size. It is therefore difficult to observe simplified predictions of growth rates according to particle size like for example the one predicted by the Kelvin effect alone. It is evident here like in other studies that nuclei mode particles below 40 nm are more hygroscopic than the immediate larger size range of Aitken particles (Holmgren et al. 2014), while for a progressive increase in size the hygroscopicity and growth factors increase accordingly.

We do refer to the chemical composition of 30 nm particles (nuclei mode), lines 290-299, but we do not state that this mode originates exclusively from traffic emissions but it may contain traffic emissions as well local and/or distant nucleating and growing nanoparticles. This is why in January and February we observe a complex state of mixing for this mode. Traffic and other combustion sources emissions may also produce particles of different chemical composition with the time scale of aging governing their hygroscopicity. This conclusion is supported by the studies we refer to for Athens and other urban areas (Wang et al., 2018; Enroth et al., 2018; Swietlicki et al., 2008) as well as recent studies (Kim et al., 2020) and the type of analysis the reviewer requires which has to do with the origin of the aerosol impacting our station. Please note that trajectory analysis by HYSPLIT may not be informative here because it has a resolution of 1^o*1^o (100 km*100km) while the size of the Athens Metropolitan area is approximately 25 km*25 km.



CPF at the 75th percentile (=2258)

We have conducted this type of analysis with local meteorological data as shown in the figure above for the 20-38 nm particle population and we conclude taking into account the map of the area that 75% of the values for this size range arrive from the center of the city. Also take into account that the distance mentioned in the manuscript between Athens city center and DEM station is around 7 km. The transport time within the Athens value at the indicative wind speeds observed are yielding estimated transport time between ½ hours to a few hours. These data provide enough evidence to assume that urban emission are the main source of these particles and adequate for aging is ensured. We would like to include this information in the supplement in order to respond in the above comments. Cluster 4 which includes Aitken and nuclei modes is a prominent size distribution pattern for 67% of the individual number size distributions. We do not find any conflict with these findings when check against individual size distributions. In any case the cluster analysis is able to distinguish the cases the reviewer is referring to. Nucleation events are very rare in the frequent heavy load aerosol present in the Athens urban atmosphere (Cluster 3, 1.4% frequency).

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.

We have to agree that cluster analysis of this very large dataset of size distributions can provide us with a qualitative picture of the dynamics and origins of size distributions in Athens. However, our findings are also based in previous papers (Vratolis et al., 2019; Bousiotis et al., 2021; Tsiflikiotou et al., 2019; Kostenidou et al., 2015) now centered and focused on the discussion regarding aerosol hygroscopicity..

We could go on a lengthy discussion if needed on the origin of these clusters and what they represent and as discussed above we have to focus on the dominant features in order to provide meaningful explanations.

For example cluster two is described as urban nocturnal because for sure it is describing traffic origin particles as indicated by the increase of its occurrence during morning hours, but it has the highest frequency during late evening hours beyond the times of evening traffic and coinciding with all traffic/residential heating and other urban emissions prevailing during the whole of the evening hours. Traffic volumes in Athens drop sharply after 9 pm at night. A discussion for the sub 10 nm particles would not be relevant here and in fact we do not use any findings from the cluster analysis for this mode resolved partly because the smaller particle size range analyzed for hygroscopicity is centered at 30 nm.

We fully agree that primary traffic particles are above 20 nm and a mixture of those plus secondary particles from condensable gaseous species is what constitutes the aerosol type dominating the 30 nm particles we analyze.

In any case we agree to reduce the current discussion on cluster analysis to maximum one page avoiding detailed descriptions of their origin and mainly focusing on the modal structure revealed with respect to the selected four sizes for the hygroscopicity analysis.

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.

All these grammar mistakes will 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.

In principle we agree to describe further this section however this will increase the length of the paper even more. We have cited the papers where this methodology is described in more detail.