Answers to reviewer comments

Anonymous Referee #1

General comments:
The paper presents long-term hygrosopicity measurements from a mountain site in Switzerland. The data is of importance as they are the first measurements of their kind to be conducted over a whole year at a site in the FT. The measurements and data analysis were additionally made in a more standardized way than previously possible. The article is well written and the results are presented in a concise manner. I recommend it for publication in ACP, however my main questions concern the interpretatition of the results as outlined in the specific comments below. These issues should be addressed before ACP.
Thank you.

Specific comments:
All the data presented in figures 2, 3, 4 and 6 are averaged over a month or more. But it is nowhere discussed how large the variation inside one month was. The annual mean of 265nm particles was 1.46, the monthly mean range is around 1.41-1.54, but the reader is left without any picture of the daily variability. For CCN activation and climate impact (which the authors also discuss), it is important to know if variation is e.g. +-0.05 or +-0.3. Please add some discussion on this.

The short-term variability of aerosol hygroscopicity is unimportant for CCN number concentration at the JFJ site. Nevertheless, it might play an important role for other processes (see also answer to comment below). Therefore we added the new Appendix A with Figure A1. We decided to provide this detailed statistical analysis in the appendix in order to keep the main story concise. Appendix A reads now:
"Here we report detailed statistics of the observed variability of aerosol hygroscopicity as some atmospheric processes may be sensitive to the short-term variability of aerosol hygroscopicity. Fig. A1 shows the mean values as well as the 10th, 25th, 50th (median), 75th and 90th percentiles of individual $\overline{GF}$ measurements (6 min values) for each month and all investigated dry sizes. Observed $\overline{GF}$ values are symmetrically distributed about their mean value for the most part, with only a small asymmetry in the 10th and 90th percentiles at the larger dry sizes. The magnitude of the variability is largely independent of season and dry size. The difference between the 25th and 75th percentile, highlighted with the colored shadings in Fig. A1, is always smaller than ΔGF=0.22 and on average between ΔGF=0.11 and 0.15 for the different dry sizes. This means that within a month 50% of all individual $\overline{GF}$ measured at a certain dry diameter fall into a narrow band with a width of $\Delta GF \approx$0.15 around the mean value. The 90th percentile is smaller than 1.60 and the 10th percentile is larger than 1.20 in almost all cases. The true variability of aerosol hygroscopicity may even be slightly smaller than reported in Fig. A1 as the measurements include additional variability due to e.g. limited number of particle counts at low aerosol concentrations. However, this effect should be small as shown by Gysel et al. (2009). A rough feeling for the variability of chemical composition can be obtained by simply assuming two component particles consisting of inorganic (ammonium sulfate) and organic material with κ values of ~0.49 (Topping et al., 2005) and ~0.10 (Gysel et al., 2007), respectively, at RH=90%. The mean values of the monthly 10th, 25th, 50th, 75th and 90th percentiles of $\overline{GF}$ at D_0=110 nm are 1.23, 1.37, 1.44, 1.50 and 1.55, respectively, which corresponds to particles consisting approximately 12%, 26%, 38%, 51%, and 62% by volume of inorganic material according to our simple model."

Topping, D. O., McFiggans, G. B., and Coe, H.: A curved multi-component aerosol hygroscopicity model framework: Part 1 - Inorganic compounds. Atmos. Chem. Phys., 5, 1205-1222, 2005.
Gysel, M., Crosier, J., Topping, D. O., Whitehead, J. D., Bower, K. N., Cubison, M. J., Williams, P. I., Flynn, M. J., McFiggans, G. B., and Coe, H.: Closure study between chemical composition and hygroscopic growth of aerosol particles during TORCH2. Atmos. Chem. Phys., 7, 6131-6144, 2007.

An underlying theme in the paper is that the authors present data over a year, show that there are differences, and finally conclude that they are within natural variations and no real trends can be found. It is not clear to me that this is the case. Additionally, if a difference in GF of 0.1 between two months is reasoned to be in the natural variability, the variability could also go in the other direction and the trend would be very strong.

More comments on this issue can be found under the specific comments.
These specific comments are addressed below.

Appendix B could be moved to online supplementary material instead of an appendix, as some of the same data is plotted in figure 5, and the data is only briefly mentioned in the article. This would make the resulting paper shorter and possibly more concise.
It is correct that the former Figures B1 and B2 (now C1 and C2) are only briefly mentioned in the paper. This is also the reason why we put them into the appendix. We keep these two Figures in the appendix for the following reasons:

- The information provided with Figures B1 and B2 is required to prove that the diurnal cycle of the overall mean GF is caused by the diurnal cycle of the mean GF of the main mode of particles with GF>1.25 rather than by a diurnal cycle of the number fraction of less hygroscopic particles with GF<1.25. This is a relevant finding which is briefly discussed in the main text and which should not be hidden in the supplementary material.
- Second, moving the Figures to the supplementary material would not make the paper more concise as they are anyway not part of the main text, except for the above-mentioned brief statement.

Page 13574, line 11: Add "the hygroscopicity parameter kappa" or similar to at least give some information what you are talking about also to readers not previously familiar with kappa.
We changed the sentence to:
"This size dependence can largely be attributed to the Kelvin effect because corresponding values of the hygroscopicity parameter κ are nearly independent of size."

13575, 23: A one month measurement can give a fairly good representation on diurnal cycles, at least for that period. The 13 months of measurement only cover one year (seasonal cycle), so using the authors definitions, even this measurement is not long enough to "provide representative information" on seasonal cycles as only one was measured.
Yes, we agree: A month-long measurement can be suitable to investigate diurnal cycles. We modified the sentence:
"Short observation periods provide only a snapshot of the aerosol properties and their diurnal cycles for the specific atmospheric transport and weather conditions encountered but they rarely provide representative information on month-to-month variations and seasonality."

13576, 23-25: Some short discussion is needed on the effect of sampling at 25C when ambient air can be 30+C lower.
Heating the aerosol from ambient temperatures to 25°C
We added the following sentences:
"Heating the aerosol from ambient temperatures to 25°C dries the aerosol and some evaporation of semi-volatile material with high vapour pressures can potentially occur. However, a comparison of parallel indoor and outdoor number size distribution measurements showed that differences in the accumulation mode could mainly be explained by evaporation of water (Nessler et al., 2003). Furthermore, previous HTDMA measurements at the JFJ performed at -10°C, 0°C and 20°C showed no substantial differences (Sjogren et al., 2008); note, these measurements were conducted in different short-term campaigns."

Nessler, R., N. Bukowiecki, S. Henning, E. Weingartner, B. Calpini, and U. Baltensperger (2003). Simultaneous dry and ambient measurements of aerosol size distributions at the Jungfraujoch, Tellus, 55B, 808-819.

13578, 8-9: The sample flow should not affect the size, only the spread. Please explain.
Yes, the sample flow rate should theoretically not affect the size selection of a DMA. However, in practise we always see some influence of the sample flow rate if the flow ratio becomes small. Therefore we have to correct for it. The effect on the width of the TDMA's kernel function was also observed, but these changes are too small to significantly affect the inverted GF-PDFs.
We changed the following sentence:
"The dilution of the sample flow drifted by ~0.2L/min during one month of operation, which offset the dry GF measurements by ~3%. The associated increase of the TDMA's kernel function width had no significant effect on the inverted GF-PDFs."

13580, 17-21: So how predominantly horizontal is the advective weather class if the mountains cause much vertical movement? Will this not impact on your analysis and conclusions?

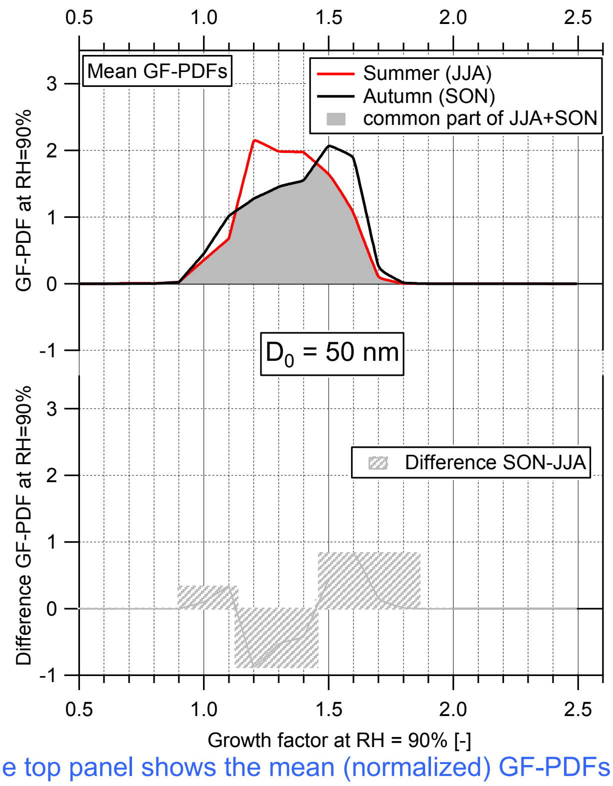
Indeed, advective weather classes can include substantial orographic lifting, though sometimes the air masses at lower levels can also move around the mountains. This has different effects, i.e. transport from the polluted Po Valley but also enhanced precipitation due to the vertical lifting. This results in counteracting effects which are already discussed in detail in Section 3.4.

13582, 2: This is a very broad range of GFs. I would like to see more discussion of these very different particle types.
The following additions have been made to the text:
"It has to be emphasized that mean GF-PDFs just represent the mean distribution of growth factors, where the appearance of e.g. a broad mode or two distinct modes in the mean GF-PDF does not imply that particles of distinctly different hygroscopicity and thus composition were simultaneously observed.
Mean GF-PDFs of D_0 = 35 nm and D_0 = 50 nm were found to be very similar (Fig. 2a and b), characterized by a broad peak within the range of 1.10<GF<1.60, indicating that the full range of compositions ranging from particles dominated by organics and/or black carbon to particles dominated by inorganic salts are present in the Aitken mode."

13582, 2-3: "Small particles efficiently acquire hygroscopic material through condensation". Compared to what? Large particles acquire material even more efficiently. Do you mean in relation to their original mass?
We clarified this sentence:
"In relation to their mass small particles efficiently acquire hygroscopic material through condensation."

13582, 3-6: "Nearby emissions of small non-hygroscopic particles are too low to show up as a distinct non-hygroscopic mode at D0=35 and 50 nm." But there are emissions like this nearby? There is also two modes in both the 35 and 50 nm total data, although this is not mentioned. The discussion on the smallest sizes should be thought through and presented better than just saying that differences are minor and due to random variations. The difference between autumn and summer/winter for 50 nm particles is so pronounced that if it is only waved of as minor, then the whole point of this study becomes questionable.
First, this sentence has been clarified:
"Absence of a distinct non-hygroscopic mode shows that nearest emissions of small non-hygroscopic particles in the diameter range between 35 and 50 nm occur too far away to retain their original properties until arrival at the JFJ."

Second, the mean autumn and summer/winter GF-PDFs at D_0 = 50 nm have much more in common than what one might think at first. This is illustrated with the following figure:



The top panel shows the mean (normalized) GF-PDFs from autumn (black line) and summer (red line) along with the common area of both GF-PDFs (grey shading). The bottom panel shows the difference between autumn and summer with equal axis scaling (i.e. area is directly comparable). This graph shows that the GF-PDFs from these two seasons have as much as 84% of their area in common, while only 16% of their area is different. The difference plot shows that the autumn GF-PDF is slightly wider with somewhat less particles in the medium GF range and a few more particles at the smallest and highest GFs. The resulting mean GFs of 1.38 and 1.34 in autumn and summer, respectively, are very similar (see Table 2). We did not add this figure to the paper but we try to acknowledge this fact with the following statement:

"Differences between seasonal mean GF-PDFs are minor and can be due to many different reasons, as will be shown later. For example the mean GF-PDFs at D_0 = 50 nm observed in autumn and summer might seem clearly different at the first glance, but they actually have 84% of their area in common, while only 16% of the particles are differently distributed with respect to GF."

Third: Distinct modes are only reported for the "strong SDE only" data, which are only a small subset of the whole data set. The appearance of these two distinct modes is not further commented because it may not be representative.

13582, 26: The fact that you see no pronounced seasonal trend of the mean GF (as mentioned also in the abstract) is perhaps true, but as discussed some lines earlier, the shape of the GF-PDF does change, and this can be very important for e.g. CCN activation.

In the response to the previous comment we have made the point that the GF-PDFs in different seasons are congruent for the most part with only minor differences. Furthermore, it has been shown that at remote sites the mixing state, which is reflected in the shape of the GF-PDF, has only a very small influence on the integrated number concentration of CCN, whereas knowing the mean GF is of importance (Ervens et al., 2010; Kammermann et al., 2010). This is due to compensating effects of larger less hygroscopic particles and smaller more hygroscopic particles.

Ervens, B., M. J. Cubison, et al. (2010). "CCN predictions using simplified assumptions of organic aerosol composition and mixing state: a synthesis from six different locations." Atmos. Chem. Phys. 10(10): 4795-4807.

Kammermann, L., M. Gysel, et al. (2010). "Sub-arctic atmospheric aerosol composition: 3. Measured and modeled properties of cloud condensation nuclei (CCN)." Journal of Geophysical Research-Atmospheres 115(D4).

I would expect more SOA to be present in particles in the summer, and more non-hygroscopic particles from increased heating or traffic sources in winter. Your data seems to indicate this (Fig. 2), but still your main conclusion is that all seasonal variations are likely only random.

Above we have made the point that the mean GF-PDFs of different seasons are largely congruent except for minor differences. Nevertheless, the hypothesis put up by the referee is indeed a possible explanation for the subtle differences between summer and winter at the larger diameters investigated. However, in summer PBL influence is observed at the JFJ for certain weather classes, while this is not the case in winter. In this sense the winter aerosol can also be considered to be more aged than the summer aerosol. Furthermore, in the discussion of Fig. 4 we point out, that more frequent Saharan dust events or occasional influence from local rock drilling works in the winter 2008/2009 might have had an influence at the largest diameter (with the most pronounced summer/winter difference). In summary, any statements about exact reasons for the subtle differences between summer and winter would be speculative.

Related slightly to the previous, later you state that kappa=0.24 is a good approximation for any model, but even the 75th percentile is already ca 0.35 for 265 nm particles. Can you still imply that kappa=0.24 for all particles is a good approximation for "any" model? Not for a model predicting CCN, at least?

The sentence in the ACPD says

"…is a good approximation in any model that needs a simple description for the hygroscopicity of the Aitken and accumulation mode aerosol …."

Models with a simple description of aerosol hygroscopicity should of course not be used to describe processes where the size dependence of aerosol hygroscopicity or the mixing state have a large influence, if computational restrictions allow to use more detailed models. Thus we believe that the sentence acknowledges the restrictions of using a constant κ (i.e. constant in time and independent of particle size) appropriately. Jurányi et al. (2010a,b) showed for example that using a constant κ value allows accurate CCN predictions at the JFJ site at any time and for a wide range of supersaturations. We have added the following paragraph to Section 3.1 (plus Appendix A with Fig. A1):

"A detailed analysis of the short-term variability of $\overline{GF}$ is provided in Appendix A and Fig. A1. Aerosol hygroscopicity was only moderately variable. 50% of individual $\overline{GF}$ measurements fall within a narrow band of ΔGF~0.15 around the median value. Nevertheless, short-term variability, seasonality, size dependence of aerosol hygroscopicity and aerosol mixing state as provided in Figs. 2-4 and Fig.A1 as well as Table 2 should be considered in models describing processes that are sensitive to small variations of aerosol hygroscopicity."

Jurányi, Z., M. Gysel, et al. (2010). "Measured and modelled cloud condensation nuclei concentration at the high alpine site Jungfraujoch." Atmospheric Chemistry and Physics 10(16): 7891-7906.

Jurányi, Z., M. Gysel, E. Weingartner, N. Bukowiecki, L. Kammermann, and U. Baltensperger (2010). 17-month climatology of the cloud condensation nuclei number concentration at the high alpine site Jungfraujoch, submitted to Journal of Geophysical Research.

13583, 6 and 13: Monthy mean values go down to 0.17, but fall within 0.3+-0.1???

We modified the sentence to

"The monthly and annual mean values of κ observed at the JFJ mostly fall within the range κ=0.3±0.1, which is the hygroscopicity value recommended for continental sites in a recent publication (Andreae and Rosenfeld, 2008)."

13583, 3: As a motivation for assuming that most differences between months are not real seasonal variations, but just natural variation, is that May 2008 and 2009 do not agree. Have the authors checked for reasons of the difference between these months? If, let's say, the difference is due to May 2008 being colder than May 2009, then the seasonality may still be a reason for the difference between summer and winter months.

Indeed we agree with the referee that a small seasonality may be present. However, many more years of measurements would be needed to extract a significant seasonality due to the natural variability of the meteorology, emissions, etc. Even in a detailed analysis we did not find a specific reason in the meteorological variables for the differences between May 2008 and 2009.

Similarly, can the high GF in September be explained by other meteorological parameters? This data should be checked, and whether or not something correlates, it should be mentioned in the text. This should be an easy task since the data is surely measured at the station

The main reason for the high mean GF in September (and in May 2008) is due to the fact that the number fraction of particles with GF<1.25 is low. Again, a detailed analysis did not reveal a specific reason in the meteorological variables for this observation.

13584, 1-7: Would it have been better to define a limit in kappa space instead of GF space? In this way the Kelvin effect would not affect the limit for the smaller particles.

We agree that using a constant κ limit instead of a constant GF limit would have been valuable alternative. Both of them have their justification depending on the process investigated. The difference between the two is actually very small for our data set. Using e.g. a limit of κ=0.1355 would correspond to GF limits of 1.22, 1.24, 1.26, 1.27, 1.28, 1.29 for the dry diameters D_0=35, 50, 75, 110, 165, 265, respectively. This does not cause much change compared to using 1.25 for all sizes because e.g. at D_0=265 nm the number fraction of particles falling between GF=1.25 and GF=1.29 is very small. We decided to use a constant limit for the primary measurement quantity (GF) rather than a constant κ limit.

13586, 21-22: You should be careful to talk about indirect climate effects based on measurements at one site. Especially when you are assessing the importance of particles formed more than 1000km away from your site.

The statement made in the ACPD paper already contains the qualifier "at this site". The fact that SDE is unimportant for the number of CCN at the JFJ site has been confirmed by Jurányi et al. (2010a). We modified the text as follows:

"As a consequence dust particles are not relevant for the total number of CCN at this site (after a transport distance of several thousand kilometers) and with that they are unlikely to have an indirect climate effect through the formation of cloud droplets. This has been confirmed by CCN measurements at the JFJ site (Jurányi et al., 2010a). However, they may still influence mixed-phase and ice clouds, if they were to act as ice nuclei"

Jurányi, Z., M. Gysel, E. Weingartner, P.F. DeCarlo, L. Kammermann, and U. Baltensperger (2010a). Measured and modelled cloud condensation nuclei number concentration at the high alpine site Jungfraujoch, Atmos. Chem. Phys., 10, 7891-7906.

13587, 6: Again, you should be very specific in the presentation when talking about climate effects of Saharan dust measured at one site in Switzerland. But I do agree on the previous two points, that you have the instrumentation and methods to assess the impact on indirect and direct aerosol effects. Only keep in mind also the geography.
We fully agree with the referee. We believe that we have taken care of the “geography” by mentioning the transport distance of Saharan dust above.

13587, 19: Are you talking about new particle formation in the FT?
This sentence has been clarified:
“By contrast, new particle formation events in the FT mainly increase n_{tot} due to high concentrations of nucleation mode particles (Weingartner et al., 1999),…”

13587, 22: Your CA weather class is defined as subsidence, and CC as lifting. It is unclear to me why subsidence leads to mixing from below, as this should be the complete opposite.
Mixing from below is due to thermally driven PBL injections during daytime. The convective class CA results in clear, sunny weather (high pressure weather situation) with very little or no advection. Especially in summer months, thermally induced vertical transport over mountainous slopes causes afternoon peak concentrations at the JFJ. Lifting and subsidence refers to large-scale air motions during the mentioned weather classes, resulting in characteristic low or high pressure situations. We clarified this in the new manuscript by adding the following sentence to the description of the Alpine weather statistics (2.4):
“Of special interest is the CA weather class. The clear, sunny weather caused by the large scale subsidence (high pressure conditions) leads in summer to injections of PBL air into the FT, locally driven by thermally induced vertical transport over mountain slopes and thus to distinct diurnal patterns with higher aerosol concentrations during the afternoon.”

13588, 5: Point out that the information is in an Appendix, unless you move it to suppl. mat.
We changed the sentences to:
“The diurnal pattern of $\overline{GF}$ is mainly caused by a decrease of the GF of the main more hygroscopic mode ($\overline{GF}_{GF>1.25}$; Appendix Fig. B1). The number fraction of less hygroscopic particles ($f_{GF<1.25}$; Appendix Fig. B2) exhibits little diurnal variation at diameters larger than 110 nm.”

13589, 4: I have not agreed with the authors on how big differences are needed to be statistically relevant, but writing that the total and FT data "are equal" is definately wrong.
We changed the sentence to:
“However, the fact that the annual mean GF-PDFs of the whole data set, and of the free tropospheric conditions only are very similar shows that the PBL influence has essentially no effect on the annual mean hygroscopic properties.”

13589, 20-25: If I understand the methodology correctly, you choose 88 (based on Fig. 5) days out of more than 200, and use this data to define that the site is representative over a regional/continental scale. And the days that are left out are the ones that by definition are most likely to not be representative. If so, then this should be pointed out very clearly also in the conclusions and abstract.
In the first paragraph of Section 3.4 we argue in detail why specifically the advective AWS classes are considered to be suitable to investigate the homo-/heterogeneity of aerosol properties on a regional/continental scale.
We changed the sentence on page 13589 to:
“For this purpose we split all data from the advective AWS classes (including more than one third of total measurement time) by the synoptic scale wind direction (north, east, south and west, as described in Table 1).”
The corresponding statement in the conclusions has been changed to:
“Aerosol hygroscopicity was found to be virtually independent of synoptic wind direction during advective weather situations, covering more than one third of total measurement time, i.e. when horizontal motion of the atmosphere dominates over thermally driven convection.”

13590, 3-4: Again, the data are not equal. As you also state later on rows 10 and 13. And I think the lower GF of 50 and 110 nm particles from south an east is large enough to mention.
”virtually equal” has been replaced, the sentence reads now:

"The integral hygroscopicity properties $\overline{GF}$ (Fig. 6a), $f_{GF<1.25}$ (Fig. 6b), and $\overline{GF}_{GF>1.25}$ (Fig. 6c) are very similar for all four synoptic wind directions at any dry size."
The differences between wind directions are already discussed with the following statement in the ACPD paper:
"The small differences in the mean GF for different wind directions correspond to a difference of as little as κ=±0.02 compared to the overall mean κ value at the corresponding dry size."

13592, 23-25: Have you not earlier stated that for example SDE:s are hardly noticed by the HTDMA, but have a large impact on scattering? Additionally, one needs to assume that kappa works perfectly (which it does not, as has been shown in other studies) to extrapolate data at 90% RH to all ambient conditions. I feel that this last sentence promises too much unless you can shortly show it in the results.
The text has been clarified:
"This data set can also be used to quantify the influence of ambient RH on light scattering by aerosol particles, which is routinely measured at the JFJ site at dry RH as shown by Fierz-Schmidhauser et al. (2010). The latter paper also describes the special treatment of SDEs."

Technical comments:
13575, 5: Remove "to that"
Done

13575, 20: Move "and references therein" after the reference
Done

13576, 18: To be clear, mention that 550nm refers to wavelength and not particle size.
We changed the sentence to
"…and the scattering coefficient at the wavelength of 550 nm, $\sigma_{sp,\,550}$, using an integrating nephelometer (TSI Model 3563) is routinely measured at this site (Collaud Coen et al., 2007)."

13577, 20: gauges
Done

13578, 20: Remove parentheses around reference
Done

13578, 25-27: The fraction is not obtained by integration, only the number.
We don't agree: The number fraction is obtained by integrating the GF-PDF (it's by definition normalized to unit area) up to the limiting GF, as written in the manuscript (see also eq. C.8 in Gysel et al., 2009).

Gysel, M., McFiggans, G. B., and Coe, H.: Inversion of tandem differential mobility analyser (TDMA) measurements, J. Aerosol Sci., 40, 134–151, 2009.

13580, 7: And CET=local time?
Yes, local winter time is equal to CET. Nothing changed.

13580, 17: are->were
Done

13581, 13-16: "Most parts of the components were stable...most parts worked reliably...". Please remove one.
We changed the sentence to:
"Only a few mechanical parts had to be replaced throughout the full year (pump membranes, smaller pumps for cooling water circulation, and humidification loop) while most parts worked reliably during the whole period (Nafion™ dryer, RH sensors, CPC)."

13581, 19-20: All these numbers are not important. It should be enough to say that 10100-10600 spectra for each dry size were measured.
We changed the sentence to:
"Between 10 100 and 10 600 valid GF-PDFs were obtained for each dry size during the 13 months of measurement on the JFJ."

13584, 11: Should be 3c and 3d?
Yes, done

13586, 5: This was investigated previously using...
Done

Fig 2 legend: What does "all seasons, oly strong SDE" mean?
This means that only data during strong SDE from all seasons were considered. This is also written in the figure caption. It reads now:
"Mean GF-PDFs for different dry sizes split by season as well as for strong SDE data from all seasons."

Fig 2 y labels: State the units.
The units are given in the bold overall label, similar to the x-labels:
"Growth factor probability distribution … 90% [-]"

Fig 3. Half the figure is left empty of data to show different legends/text boxes. This makes the data hard to read. The legend can be made 2x3 instead of 6x1, and the text boxes can be at least decreased in size.
We re-arranged the D_0-legend. However, the scales of the y-axes were on purpose chosen to cover equivalent ranges. E.g. the growth factor range GF=1.25-1.70 corresponds approximately to the κ range 0.1-0.6 and covers more or less the full range of mean growth factors / κ values observed in continental air masses.

Anonymous Referee #2

The paper presents an extensive record of aerosol hygroscopicity observations from the Jungfraujoch site, a globally important site for climate relevant studies. The amount of data contained in this analysis is really quite impressive, as is the feat of maintaining this measurement campaign over the year plus timeframe. My compliments to the authors on this undertaking. The paper clearly written and presented, albeit somewhat length. The measurements were completed with requisite care and due diligence. A few improvements can be addressed before this is published.
Thank you

The authors provide the best information on aerosol hygroscopicity for a free Tropospheric site which will be clearly useful for the modeling community. The seasonal consistency of the hygroscopic properties is certainly notable. Really the largest perturbation is the diurnally timed transport of PBL air masses, quite interesting. This is certainly a take home message of the analysis and should be emphasized.
We believe that the following statement in the abstract is clear enough:
"Thermally driven injections of planetary boundary layer (PBL) air, particularly observed in the early afternoon of summer days with convective anticyclonic weather conditions, lead to a decrease of aerosol hygroscopicity. However, the effect of PBL influence is not seen in the annual mean hygroscopicity data because the effect is small and those conditions (weather class, season and time of day) with PBL influence are relatively rare."

At the same time, I agree with the first reviewer's comments directed at what really constitutes a significant seasonal, diurnal, dry size, meteorology class etc. variation. What might help here is a summary table (or added into table 2) an expression of the variability in a consistent metric (e.g. Coefficient of Variability). It will be clearer to see what is important and in what context a 'universal' constant kappa value can be applied without concern over the impacts. Is there a gauge given (e.g. by Petters et al.) as to what variations in kappa are of significance? Defining somehow what constitutes a significant variation in kappa or GF would help here. Overall, I do agree with the general conclusions of the general lack of strong variation as a function of season, dry size, meteorology class, but it just must be better quantified/substantiated.
The first reviewer's comments are addressed above. If a certain variation in κ has significant effects depends on the process that is investigated. As an example it will be different for direct and indirect effects. Therefore it is up to the user to determine what degree of simplification is possible in a specific application. E.g. Jurányi et al. (2010a) provide a detailed sensitivity analysis of CCN concentrations to variations of κ.

Jurányi, Z., M. Gysel, et al. (2010a). "Measured and modelled cloud condensation nuclei concentration at the high alpine site Jungfraujoch." Atmospheric Chemistry and Physics 10(16): 7891-7906.

Figure 2 takes a lot of looking to see which seasonal trace is which. Can it be improved? I don't see where it would help though to break this out into 4 times as many plots with each season on a separate panel. The point is the seasonal variation is not so important so leaving it as such is merited.
Figure 2 was not changed as eventually suggested.

I would move the first paragraph of the results section to the instrument section of the paper.
We moved the first paragraph of the results section to the very end of *2.2 Instrumental*

Beyond these comments and the details covered by the other review, I believe this paper is ready for publication.
Thank you!

Anonymous Referee #3

Overall this is a very good paper that addresses hygroscopic properties of atmospheric aerosol. This is a property that is of consequence to global modeling and radiation transfer and climate science. The results presented are representative of the regional scale of Western Europe. They include seasonal and synoptic variability as well as shorter term variability and quantitative presentation of this variability in addition to the central values of hygroscopcity.
More emphasis should be placed on the variability and its importance as input to models.
Thank you

Page 13577
"Thereafter, a monodisperse size cut with dry diameter D0 is
selected using a differential mobility analyzer (DMA) and subsequently humidified by transferring water vapor through a Gore-Tex™ tube into the sample flow."
Suggest, "size increment" or rather than " size cut".
Done

Page 13578
"All data measured at lower or higher RH were ignored."
Suggest, "All data acquired at lower or higher RH were ignored."
Done

"Different integral properties . . . "
I would call these central values of the PDF rather than integral properties.
We believe that "integral properties" is a suitable term, since $\overline{GF}$, $\overline{GF}_{GF>1.25}$, $f_{GF<1.25}$ are values that are obtained by integrating the GF-PDF (or a subrange thereof) with a weighting specific for each quantity (detailed equations can be found in Appendix C of Gysel et al., 2009). The term "central values" rather refers to a median value to our understanding. Nothing changed.

Gysel, M., McFiggans, G. B., and Coe, H.: Inversion of tandem differential mobility analyser (TDMA) measurements, J. Aerosol Sci., 40, 134–151, 2009.

Page 13579
"Two particles of different size but identical chemical composition will
have the same because the influence of the Kelvin effect is filtered."
Suggest, ". . . .because the influence of the Kelvin effect is removed according to equation 2, below".
Done

"We assumed surface tension of pure water in our calculations."
What error is associated with this assumption?
HTDMA-derived κ values are sensitive to including the Kelvin term in the calculations (1st order effect) but they are rather insensitive to the exact value used for the surface tension (2nd order effect). The following sentence has been added to the manuscript:
"HTDMA-derived κ values are only weakly sensitive to the exact value of the surface tension. If the true surface tension was e.g. 10% lower than that of pure water, then κ would be overestimated by only ~1.2% at D_0=110 nm."

Page 13580
"During winter this effect is hardly found, and the air masses present are usually representative of the free tropospheric background conditions (FT)."
Suggest, "During winter this effect is found infrequently,. . . ."
If this can be quantified by percentage of time observed then all the better.
The sentence was adapted as suggested by the referee.
Accurate distinction between FT and PBL-influenced-conditions requires a thorough analysis of numerous gas phase and aerosol parameters for each day, which is difficult for a 13-months data set and out of the scope of this study.

Page 13581
"A total of 10 408, 10 575, 10 626, 10 596, 10 532, and 10 115 valid GF-PDFs were obtained for D0=35, 50, 75, 110, 165, and 265 nm, respectively, during the 13 months of measurement on the JFJ."
Simplify to "Approximately 10,000 . . . .. were obtained at each Do value during . . . .."
This sentence was simplified (see also comments by referee #1, above)

Explain why the N for the largest Do is significantly different if this is of importance with respect to your results and aerosol properties, and continue with the next paragraph.
The reason for this is that towards the end of the campaign discharges occurred in DMA2 during some of the 265nm-scans. Such data had to be discarded. The remaining valid data are unlikely to be biased in some way as only a small fraction of the data was invalid.

"Seasonal mean GF-PDFs were calculated by averaging these individual GF-PDFs separately for each season and dry size (Fig. 2a–f). It has to be emphasized that mean GF-PDFs just represent. . . "
"Separately" and "just" not needed.
"Separately" and "just" was removed.

Page 13582
". . . of small non-hygroscopic particles are too low to show up as a distinct nonhygroscopic . . . "
Are the emissions to low or the concentration of gas or particulate phase hygroscopic mass too large and accumulation of hygroscopic mass too fast (efficient) as in previous sentence?
This paragraph was modified (see answer to comments by referee #1, above)

"They are characterized by a dominant contribution of more hygroscopic particles with growth factors between 1.25–1.7, as well as a minor contribution of less hygroscopic particles with growth factors .1.25."
As stated earlier, the averaging precludes that there were ever two separate modes in the GF PDF. Were there significant occurrences of two modes simultaneously? To show this would require a different statistic.
Indeed, our analysis does not show whether two or multimodal GF-PDFs occur frequently (and this point is made clear in the manuscript). It would of course be interesting to address this question in more detail. However, aerosol number concentrations at the JFJ are very low such that a more detailed analysis of the exact shape of individual GF-PDFs is only possible for times and sizes with highest aerosol number concentrations (Gysel et al. (2009) discussed the limitations imposed on the interpretability of TDMA measurements taken under conditions with low aerosol number concentrations in detail). Thus we do not present a more detailed analysis, which could only be done for a small and potentially non-representative subset of the whole data set.

Gysel, M., McFiggans, G. B., and Coe, H.: Inversion of tandem differential mobility analyser (TDMA) measurements, J. Aerosol Sci., 40, 134–151, 2009.

"The annual cycle and the size dependence of integral properties of the GF-PDF are discussed in the following."
This does not add much to the discussion; it is basically the section heading repeated.
We skipped this sentence.

Page 13583
"Monthly mean -values, determined from the observed hygroscopic growth according to Eqs. (1) and (2), varied in the narrow range between 0.17 and 0.31 . . . "
To say that the range is narrow requires some reference either internal from your own data set or some other set, atmospheric or otherwise.
We modified the sentence:
"…varied in the range between 0.17 and 0.31 and were similar for all…"

Similarly, in connection with this and results in related paragraphs, to state that ". . . no pronounced seasonal trend was found for GF. . . "
one should discuss the importance of variation and standard deviations for the average monthly values or the uncertainty in the g(RH) and _galues.
The new Figure A1 quantifying the short-term variability of the hygroscopicity was added to the paper (see also answer to comment by referee #1).

While a constant k value is convenient and maybe OK for a simple description of hygroscopicity most models can handle seasonal and synoptic variability. I would not want modelers to take this as regionally and seasonally constant. Thus, the inter and intraseasonal distribution of GF as presented in Table 2 should be pointed out to the reader. What effect would the range or uncertainty of kappa have on say CCN potential at low supersaturations or on extinction, direct radiative effect, for a given RH?

This has been addressed with our answer to a similar comment by referee #1.

Page 13584
"In a further analysis separated mean GF-PDF. . . ."
In a further analysis, separate mean GF-PDFs. . . .
Done

GF-percentiles are equivalent to a simplified GF-PDF because each GFpercentile corresponds the a single point of the cumulative distribution function.
.. corresponds to the. . . .
The sentence reads now:
"…corresponds to a single point of the cumulative…"

Page 13587
. . . .can be explained by the fact that the dust particles are not very numerous but non-hygroscopic and predominantly in the coarse mode size range (see Fig. A1), with over-proportionally large scattering cross section.
Awkward. I suggest,
. . . can be explained by the fact that although the dust particles are not very numerous, they are non-hygroscopic and predominantly in the coarse mode size range (see Fig. A1).
Because they are larger they have a more dominant effect on scattering cross section relative to the FT or PBL accumulation mode at other times.
Done.

"Here we investigate the diurnal patterns of the hygroscopic growth factors for different weather classes. . . "
Here we investigate the diurnal patterns of the hygroscopic growth factors for routinely determined weather classes at JFJ. . .
Done.

In Fig. 5 legend, spell out the weather classes and acronyms, e.g., Convective Cyclonic, CC as in section 2.4
Done. Figure 5 and similar appendix figures were modified.

"The diurnal pattern of GF spans values from 0.33–0.15 and 0.27–0.20"
"The diurnal pattern of GF spans values from ca. 0.33 to ca. 0.15 and 0.27 to 0.20
The summary paragraphs of section 3.3 are largely redundant with the descriptive paragraphs and should be integrated or eliminated.
The sentence was modified to
"The diurnal pattern of $\overline{GF}$ spans κ values from ca. 0.33 to ca. 0.15 and ca. 0.27 to ca. 0.20 at D_0=35nm and 265nm, respectively,…"
The summary paragraph in Section 3.3 is meant to be a conclusive repetition of the key findings discussed before. We prefer to leave this structure.

Page 13590
". . . Fig. 6 did not change anything."
Fig. 6 did not change the results.
Done.

Page 13591
"Consequently a constant and size independent value of 0.24 is a good approximation in any model that needs a simple description of the Aitken and accumulation mode aerosol at the JFJ site."
Don't underrate your results to a single average number for use in models with simple hygroscopicity parameterization. The real value here is that you have measured values of kappa or GF that are regionally representative and that show seasonal and synoptic variation that is of significance when put in the context of direct radiative forcing or potential CCN activity.
This has already been addressed with answers to above comments.