We thank reviewer 1 for his helpful comments and suggestions. We have addressed the comments point by point below.

Minor comments:

Comment 1: Page 6769: If local home heating devices contribute to aerosol with very low GF values (1.0-1.1) when the site is influenced by the PBL in winter, then what fuel is likely to be used? Biomass burning aerosol is associated with much higher GF values earlier in the text, presumably if the inorganic aerosol content is high, or if secondary inorganic ions are internally mixed. The very low GF values observed here point to either BC-rich biomass burning aerosol or BC-rich particles associated with combustion of different fuel. Some discussion of potential heating fuels would be useful here. Referring to previous SP2 measurements of fossil fuel and biomass burning plume BC mode diameters may also be useful, for example (Schwarz et al. 2008). Furthermore, if the seasonality controls the PBL intrusions, isn't it possible that more fossil fuel combustion aerosol, associated with traffic, for example, is observed in the winter because of the PBL effect.

Reply 1: On page 6766, lines 16-18, we state that BC and mineral dust has a GF of less than 1.05, while biomass burning shows a large range of GF that can go up to 1.65. We now clarify in the text that *"These very high values were measured after the biomass burning particles have had some time to mix with secondary inorganic ions, and much lower GFs were measured closer to the source".*

According to the referee comment, it has also been added to page 6769 that *"The very low GF values observed here point to either BC-rich biomass burning aerosol or BC-rich particles associated with other combustion sources, such as fossil fuel combustion aerosol, which have had little time to mix with secondary inorganic ions."*

Moreover, we clarify the role of the seasonality of PBL intrusions on the seasonality of the hygroscopic behaviour of particles (see answer to reviewer 2's comment no. 9).

Comment 2: Page 6770, first paragraph: Particles with relatively high GF values may still contain BC. If the inorganic ion volume fraction is very high, it is not possible to assess whether a small BC core is or is not present using this method.

Reply 2: Yes, this is true. We have now mentioned this in the text: *"It should also be mentioned that particles with relatively high GF values may still contain BC if the inorganic ion volume fraction is very high; it is not possible to assess whether a small BC core is present using this method."*

Comment 3: Page 6773: Is it possible that partitioning of nitrate or ammonium nitrate at lower temperatures at night also contributes to the higher night time mean GF values?

Reply 3: Yes, it is indeed possible and we now mention this possibility in the text: *"It is also possible that the partitioning of nitrate or ammonium nitrate at lower temperatures contribute to the higher GF values at night."*

Comment 4: Page 6775, line 1: Do the authors mean that the primary marine particles are internally mixed? Displacement of sea salt chloride by nitric acid during mixing with anthropogenic plumes? Or do sea salt particles have a minimal contribution to number at 165 nm? A GF of 1.8 would be too high for an OA-ammonium sulphate internal mixture, but it may be reasonable for a particle composed predominantly of sodium nitrate.

Reply 4: A GF of 1.8 would indeed be reasonable for aged sea salt particles, either by displacement of sea salt chloride by nitric acid, or by mixing of NaCl with less hygroscopic species such as organics, ammonium sulphate or ammonium nitrate. We now precise these possibilities in the text: *"A GF of 1.8 can represent aged sea salt particles, either by displacement of sea salt chloride by nitric acid, or by mixing of NaCl with less hygroscopic particles such as ammonium sulphate or ammonium nitrate (Gard et al., 1997)."*

Comment 5: Fig. 10 caption: How is this seasonal variation? Is this not classified based on air mass origin?

Reply 5: This is a correct observation; the caption is wrong and has been changed.

Comment 6: Fig. 11 is perhaps not necessary as the GF depends more on season than air mass origin as discussed in the text.

Reply 6: We believe that Figure 11 is necessary, as we show by splitting seasonal variations by air mass types, that the general seasonal variation (without air mass splitting) is not observed in individual air mass types. In fact both the seasons and air mass type influence the hygroscopic properties of the aerosols at puy de Dôme.

Specific comments:

Comment 7: Page 6765: Should be "Matlab"

Reply 7: Lines 10, 16 and 18 - Yes, this has been changed.

Comment 8: Page 6766: should be "biomass burning aerosol"

Reply 8: Line 18 – Yes, this has been changed.

Comment 9: Page 6766: Duplissy et el. 2011 observed higher GF values for aged SOA in simulation chamber studies (Duplissy et al., 2011)

Reply 9: Line 21 –This information, and the reference, have been added. *"GFs of secondary organic aerosol (SOA) (...) and up to more than 1.65 for aged SOA in simulation chamber studies (Duplissy et al., 2011)."*

Comment 10: Perhaps shorten the discussion around Fig. 11. The main point is that there is a dependence upon season but not necessarily air mass origin.

Reply 10: see previous reply to comment 6.

References:

Duplissy, J., DeCarlo, P. F., Dommen, J., Alfarra, M. R., Metzger, A., Barmpadimos, I., Prevot, A. S. H., Weingartner, E., Tritscher, T., Gysel, M., Aiken, A. C., Jimenez, J. L., Canagaratna, M. R., Worsnop, D. R., Collins, D. R., Tomlinson, J., and Baltensperger, U.: Relating hygroscopicity and composition of organic aerosol particulate matter, Atmos. Chem. Phys., 11. 1155-1165, doi:10.5194/acp-11-1155-2011, 2011.

Gard, E. E., Kleeman, M. J., Gross, D. S., Hughes, L. S., Allen, J. O., Morrical, B. D., Fergenson, D. P., Dienes, T., Gälli, M. E., Johnson, R. J., Cass, G. R., Prather, K. A.: Direct observation of heterogeneous chemistry in the atmosphere, Science, 20, 1184-1187, doi: 10.1126/science.279.5354.1184, 1997.

Schwarz, J. P., Gao, R. S., Spackman, J. R., Watts, L. A., Thomson, D. S., Fahey, D. W., Ryerson, T. B., Peischl, J., Holloway, J. S., Trainer, M., Frost, G. J., Baynard, T., Lack, D. A., de Gouw, J. A., Warneke, C., Del Negro, A.: Measurement of the mixing state, mass, and optical size of individual black carbon in urban and biomass burning emissions, Geophys. Res. Lett., 35(13), L12810, 2008.

We thank reviewer 2 for his numerous comments and remarks which greatly helped to improve the manuscript. We have done our best to follow the suggestions made, and our answers are reported below.

General comments: The authors present humidified tandem differential mobility analyzer (HTDMA) measurements and hygroscopic growth factors (GF) for aerosol sampled at the French high-altitude research station, Puy de Dome. Measurements are reported for September of 2008, December-May 2009, September-February 2010, and continuously from January 2011 to December 2012. The state goal of this paper is to explore how aerosol hygroscopic growth factors vary year-to-year, seasonally, diurnally, and with air mass type. Given the incomplete dataset for 2008-2012 it is hard to draw definite conclusions regarding whether the observed differences in GF are due to seasonally-varying aerosol characteristics or due to an analysis approach biased by inclusion or lack of data for a given season in a given year. A revised manuscript should limit the scope of the dataset to ensure an unbiased analysis. In addition, a number of speculative conclusions are drawn from the HTDMA data regarding aerosol composition and emissions source attribution that are currently not supported (some instances noted under specific comments below); if these observations are based on additional measurement data, they should be discussed more fully or otherwise removed. Finally, this manuscript would be greatly improved by including all of the measured HTDMA sizes (given the non-linear nature of the observed hygroscopicities) and by a more extensive and sophisticated discussion of aerosol mixing state that incorporates the GF "spread" of each mode.

Reply: Regarding the non-continuous aspect of the data set; previous to the publication of the paper in the present form, we actually did an analysis of the seasonal variation of hygroscopic aerosol properties over the time period ranging from January 2011 to December 2012, excluding the years 2008, 2009 and 2010, which were less complete. The average seasonal variation obtained using this restricted time period were very similar to the one obtained with the whole data set (and also undistinguishable from the seasonal variation obtained over the years 2011-2012), so we decided to keep all data in the submitted version. However, we agree that we can not drive robust conclusions on a year-to-year variability when using 2008, 2009 and 2010 that do not contain sufficient data. Consequently, we have modified the figures and text of the paper excluding the 2008-2010 period regarding the year-to-year variability. Because the year to year variability is shown to be small, we will keep them in the analysis when discussing the air mass type dependency, and all analysis that are split by season.

Regarding the number of studied particle diameters; we will follow the reviewer recommendation of showing the behaviour of all diameters in the Su et al. (2010) manner in the first general figures showing yearly and seasonal averages. (see also comment and reply 3). However, for the mixing state quantifications and air mass analysis, the paper would be too heavy if we showed the behaviour of all diameters, and we wish to continue showing the behaviour of particles whose diameters are representative of the nucleation, Aitken and accumulation modes usually observed at the puy de Dôme station (Venzac et al. 2009).

Finally, the GF spread information is already included in our analysis, by the fact that when the GF spread is higher than expected ($\sigma > 0.10$) from the DMA transfer alone, two different hygroscopic modes are searched to explain this spread. This is similar to the analysis by Sjögren et al. (2008), who suggested the presence of two modes when the spread in the GF-PDF was greater than 0.15. The aspect of this type of analysis, with a strict mode fitting procedure, enables a more accurate idea of the aerosol mixing state. We have stressed this point in the analysis section of our paper.

Specific comments:

Comment 1: HTMDA measurements were made at PdD at six sizes: 25, 35, 50, 75, 110, and 165 nm, but only measurements for three sizes (25, 50 and 165 nm) are reported. Given the similar GF-derived kappas reported for 25 and 50 nm particles in Table 7 and the much higher kappa reported for 165 nm, there is clearly a non-linear size-dependence to kappa. Thus, it is essential that the

authors add the other 3 measured diameters to the manuscript in order to provide greater size resolution in understanding how the aerosol composition is varying.

Reply 1: See the answer to this in the general comment above. We know include two additional figures of kappa PDF as a function of particle size for all particles measured (see comment and reply 3). A small section is added to section 3: *"In the first stage of data analysis, the general behavior of the hygroscopic properties of particles is studied as a function of all sampled particle sizes (25, 35, 50, 75, 110 and 165 nm). Then, focus is put on the analysis of the hygroscopic growth and mixing state of particles with dry sizes 25, 50 and 165 nm. These sizes of aerosol particles are representative of the nucleation mode (25 nm), the Aitken mode (50 nm), and the accumulation mode (165 nm) at the pdD (Venzac et al. 2009). "*

Comment 2: In section 3.1 and Figures 2-7 it does not seem appropriate to include a single month as the annual average for 2008 and half a year for the annual average in 2009 and 2010. This only leaves two years for assessing "year-to-year" annual average variability, which obviously cannot lead to very robust conclusions. It would make sense to me to focus on reporting average aerosol properties over the continuous 2011-2012 time period rather than trying to identify an inter-annual trend from this dataset. Similarly, the "unbalanced seasonal sampling" identified on Pg. 6767, Lines 17-18 is a major flaw in this analysis that it must be corrected, perhaps by again focusing on the only two continuous years 2011-2012 or by bringing in only the winter data from 2009-2012. As it currently stands, definitive conclusions cannot be drawn from regarding GF month-to-month variability.

Reply 2: The year-to-year analysis now only includes the continuous sampling years 2011-2012.

Comment 3: Given the large amount of size-resolved GF data being compared in the tables and in Figures 2 and 4, I recommend that the authors report the data using the hygroscopicity distribution concept as described by Su et al., Atmos. Chem. Phys., 2010 (doi:10.5194/acp-10-7489-2010). Cumulative H (kappa, Dd) vs. Kappa curves facilitate the same comparisons but would also more easily convey the contribution of non-hygroscopic aerosols as well as the means and spreads of the hygroscopic modes. Similarly, since there are six different HTDMA sizes, interpolated color maps similar to those in Figure 9 of Su et al. would be an especially nice way to display this dataset.

Reply 3: Figures of aerosol hygroscopicity (GF and kappa) as a function of size, similar to those presented in Su et al., 2010, now complete figures 2 and 4, and text has been added to section 3.1:

"The size dependent hygroscopicity solely due to the aerosol composition can be addressed by plotting κ-PDF as a function of the aerosol diameters, calculated for 25, 35, 50, 75, 110, and 165 nm (Figure 2b) using the hygroscopicity distribution concept as described by Su et al., 2010. Both in 2011 and 2012, the aerosol hygroscopicity is increasing with increasing particle size independently of the Kelvin effect from 50 nm to larger sizes, There is also a tendency of the nucleation mode particles to be slightly more hygroscopic than the Aitken mode particles. Years 2011 and 2012 are on average very similar, with more dispersion in the hygroscopicity of the accumulation mode particles during 2012."

"*Again, the hygroscopic properties of aerosol solely due to their chemical composition can be investigated using the κ-PDF as a function of particle size. Figure 4b indicates that the higher hygroscopicity of nucleation mode particles relative to the Aitken mode particles observed on the yearly average is mainly seen in autumn and winter. Otherwise the particle hygroscopicity, as well as the external mixing degree, is increasing with particle size for all seasons. The appearance of a more hygroscopic and a less hygroscopic mode at larger particle sizes is mainly observed during autumn and winter.* "

Comment 4: I'd like to see a much more extensive discussion of aerosol mixing state beyond the current method, which defines an external mixture as when two or more modes are present. For example, the spread of each mode also contains information about mixing state, and should be more fully discussed. In the end how representative is an average GF reported in these tables (and those in other studies) of the individual aerosol particles typically present at this site?

Reply 4: See answer to the general comment. This information contained in the spread of each mode is taken into account by our method. We discuss the frequency with which more than one mode is observed (determined also from the GF spread), but also quantify the number fraction in each mode.

Comment 5: Please add kappa axes to Figures 1-7, 10-11 that correspond to the GF axes so that the reader is able to easily see the compositional differences across aerosol sizes without having to account for the (admittedly small) Kelvin size dependence of the GF.

Reply 5: Kappa axes have been added to the Figures. A short description on how kappa was calculated has been added to section 2.3.1: *"Further, hygroscopic kappa values κ, as introduced by Petters and Kreidenweis (2007), are calculated according to Eq. (1)*

$$\kappa = \frac{(GF^3-1)(1-a_w)}{a_w}, \qquad (1)$$

where a_w is the water activity, at which the GF was measured. According to Köhler theory (Köhler, 1936), a_w is obtained by Eq. (2)

$$a_w = \frac{RH}{exp\left(\frac{4\sigma_S v_w}{RTD}\right)}, \qquad (2)$$

where σ_s is the surface tension of the solution droplet (here assumed to be pure water), v_w is the partial molar volume of water in solution, R is the universal gas constant, T is the temperature, and D is the diameter of the droplet. "

Comment 6: Pg. 6762, Line 25-27: This statement is untrue and should be stricken. See for example, Sjögren et al., 2008, which is cited in the previous paragraph, who made measurements at Jungfraujoch during multiple, 1-month, campaigns spanning between different seasons in 2000-2005. Kammerman et al., 2010 (also cited in the previous paragraph) made measurements at Jungfraujoch for a continuous 13-month period.

Reply 6: The sentence has been changed to "*This is, to the authors' knowledge, the first study that investigates hygroscopic properties measured at a high altitude site almost continuously over more than two years".*

Comment 7: Pg. 6763, Lines 5-6: I don't understand the meaning of the definitions "background site" and "high altitude site" in the context of the citations and the HTDMA measurements. A description of what characteristics go into these labels would be preferable.

Reply 7: Asmi et al. 2011 define a high altitude site as a site at more than 1000 m a.s.l, and place Puy de Dôme in that category. "Background site" should be "rural background site", which is defined as site with a distance 10-50 km from large pollution sources by Putaud et al. 2004. This has been corrected in the text. The reference Venzac et al. 2009 is removed. It has also been added that *"According to Henne et al. (2010), the site is representative of western European air masses over a large scale."*

Comment 8: Pg. 6764, Lines 15-19: I don't understand how the authors are calibrating their DMAs using ammonium sulphate. I would think that this is not really a calibration, but rather a consistency

check of the instrument operation using a known pure-component salt. This should be removed or, if I'm mistaken and a calibration was performed, more details are needed.

Reply 8: We are actually evaluating the accuracy of the RH sensors in the HTDMA. Since we know at a given RH how much ammonium sulphate should grow, we adjust the real RH attributed to the measurement method using this in the Gysel software. This has been clarified in the text: "*The accuracy of the RH sensors of the HTDMA at the pdD is regularly checked using pure ammonium sulphate, which has a well known GF value at 90% RH. Intercomparisons performed during a EUSAAR workshop (Duplissy et al. 2009) showed that the average variability of measurements compared with other HTDMA instruments is less than 1%. The humidification system presented a stability of ±3% RH in normal conditions. "*

Comment 9: Pg. 6769, Lines 16-22: This result is surprising since I might expect shallow boundary layers during winter to prevent PBL air from reaching the high altitude site. Also, please provide support for the claim that "the nearly hydrophobic mode observed in winter and autumn originates from the presence of combustion aerosols emitted from heating devices".

Reply 9: During the winter time the boundary layer height can raise over the height of the puy de Dome station bringing within it large quantities of anthropogenic aerosols that have been concentrated in the shallow boundary layer. These events are isolated and are short-lived, but are responsible for the hydrophobic mode observed during the winter. This is now better explained in the text, and we bring supporting information on the presence of higher concentrations of organics and nitrates are measured during the winter months at the puy de Dome site and also a higher fraction of biomass burning aerosol during the winter at the site:

"Furthermore, higher fractions of biomass burning aerosol have been identified at the site during (Crippa et al., 2014). Asmi et al. (2012) measured aerosol cloud activation at the pdD and report that aerosol properties, and likely also their sources, are different between summer and winter. During summer, the authors measured less aged organics, possibly from biogenic sources. Winter time organics had a higher organic aerosol m/z ratio to total organics (f44), suggesting more aged organics."

We can't provide support for the statement that "the nearly hydrophobic mode observed in winter and autumn originates from the presence of combustion aerosols emitted from heating devices", so this sentence has been removed.

Comment 10: Pg. 6770, Lines 19-22: Is there any evidence to support the statement: "in autumn and winter, the high degree of external mixing may also be explained by more combustion aerosol being injected high in the atmosphere due to the very strong convection in the warm outflow".

Reply 10: We have only visual evidences of many wood fires during autumn and the beginning of winter. We have removed this statement.

Comment 11: Pg 6772, Lines 1-18: Is this discussion regarding seasonal differences in BC or in regional biomass burning supported by any measurements or observations from the site. It is mentioned on Pg. 6763, Line 10 that particulate BC, NOx, and CO2 are measured at the PdD during the study period; are these combustion tracer measurements consistent with the discussion in this section over the HTDMA measurement time period? What is the observational evidence for the statement: "In winter and in PBL conditions, the contribution from biomass burning is greater than in summer, bringing with it a high fraction of primary organics and nitrates"?

Reply 11: Higher concentrations of organics and nitrates are measured during the winter months at the puy de Dome site (Freney et al., 2011) and also a higher fraction of biomass burning aerosol have been identified during the winter at the site (Crippa et al., 2014). See reply to comment 9.

Comment 12: Pg. 6772, Line 25 and discussion throughout: How is the height of the PBL or RL being assessed so as to apportion specific aerosol types as deriving from the PBL or the FT? Presumably, this is coming from the vertical component of the HYSPLIT back trajectories or possible sonde-based temperature profiles?

Reply 12: A small section has been added to the methods/site description section (2.1) of the paper: "*Boundary layer heights (BLH) over several seasons have been calculated from model outputs (Venzac et al. 2009) or retrieved from remote sensing measurements (Boulon et al. 2011). BLH retrievals were cross-checked using meteorological parameters (Boulon et al. 2011) and in situ aerosol measurements (Venzac et al. 2009). Both studies indicate that in winter, spring, and at night during summer, the site is more often influenced by FT or residual layer (RL) air masses, compared to the summer season (Venzac et al. 2009; Boulon et al., 2011).*"

Comment 13: Pg. 6773, Line 5-8: It should be easy to detect periods influenced by NPF by examining the variation in the overall particle number concentrations referenced on Pg. 6773, Line 10. It would be very interesting to know how frequently NPF events were observed during this study period in explaining the HTDMA results.

Reply 13: The frequency of NPF events has already been mentioned in the text, pg 6773, lines 11-15, that: *"In winter, the day time increase in the number fraction of less hygroscopic particles (NF1) is smaller than during the other seasons. This is consistent with observations by Rose et al. (2013), who report that NPF events are less frequent in winter (NPF event frequency 17%) than in spring, summer and autumn (NPF event frequency 26%, 27% and 24%, respectively)."*

Comment 14 Pg. 6774, Line 2-4: I don't understand the connection between lower wind speeds and a higher PBL.

Reply 14: We only point to the fact that within the PBL, wind speeds are lower than in the FT. Hence, when the site is within the PBL, air masses are expected to be more local than when it lays in the FT.

Comment 15: Pg. 6774, Lines 15-21: I'm surprised that the HTDMA measurements at 25-265 nm would be substantially influenced by dust, which tends to be present at larger particle sizes (>~0.5-5 micron diameters).

Reply 15: Sjögren et al. (2008) report changes in the GF-PDF mainly for particle size 250 nm, and Van Dingenen et al. (2005) observed a strong less hygroscopic mode during dust events for particle sizes 100 and 200 nm. We only cite the results. This has been corrected in the text: *"Sjögren et al. (2008) report this behaviour for 250 nm particles, and Van Dingenen et al. (2005) for 100 and 200 nm particles. During dust events, there was an increase of non-hygroscopic particles at around GF 1.0."*

Comment 16: Pg. 6775, Lines 6-8: What is the basis for the statement that anthropogenically-influenced marine aerosol is more "aged" than those originating from the continental PBL?

Reply 16: This sentence was unclear and has been removed.

Comment 17: Pg. 6775, Lines 21-25: I don't understand the formation mechanisms for "freshly formed anthropogenic aerosol" and "aged anthropogenic aerosol". What type of particles are these – organics and sulphate? "Aging" of fresh combustion particles is mentioned in this sentence as well.

Reply 17: By "freshly formed anthropogenic aerosol", we mean fresh anthropogenic emissions (BC and primary organics), and by "aged anthropogenic particles" we mean secondary organic and inorganic compounds. This has been clarified in the text: *"Here, the less hygroscopic aerosol particles are likely originating from fresh anthropogenic emissions (BC and primary organics), and the hygroscopic mode corresponds to aged anthropogenic particles (secondary organic and inorganic compounds)."*

Comment 18: Pg. 6776, Lines 10-12: The meaning of the sentence "Continental aerosol displays the same trend as smaller particles, with higher hygroscopicity in the cold season" is unclear.

Reply 18: The sentence has been clarified: *"Continental accumulation mode particles display the same seasonal trend as smaller particles (nucleation and Aitken), with higher hygroscopicity in the cold season."*

Comment 19: Section 3.4: I don't understand the reason for using the Zhou et al., 2001 parameterization since it is not physically based and because it obscures the inherent size-dependent measurement variability by reducing the data to two arbitrary coefficients. I recommend cutting this parameterization and only including the kappa values as is currently done while also adding their standard deviation. In addition, the hygroscopicity distribution concept of Su et al., 2010 seems perfect for describing this dataset, while preserving the spread of the GF distribution and the frequency of observation.

Reply 19: We believe that using the Zhou et al. (2001) parameterization is straightforward for users who are not necessarily familiar with the kappa theory. Users can directly grow their aerosols in a realistic way, according to air mass type and season. We believe that this parameterisation is useful (and harmless in the paper) and would like to keep it.

Comment 20: Pg. 6777, Lines 13-18: The statement, "The parameterization and the kappa values are issued from a long term data set, and are therefore reliable for future use in studies in which the hygroscopic properties should be taken into account, such as for calculations of condensational sink or for calculation of size distributions at ambient humidities for calculations of optical properties. The parameterizations and kappa values are representative of western European aerosol in remote sites", is unfounded and should be removed.

Reply 20: The pdD site has been shown to be representative of western European air masses over a large scale (Henne et al. 2010) and we do not believe that this statement is unfounded.

Comment 21: Pg. 6778, Lines 10-11: What is the basis for this statement regarding ageing of fresh continental combustion aerosols? Is this based on elevated BC measurements?

Reply 21: We are clearly saying that we are speculating. We will remove this sentence.

Comment 22: Figure 1: How were the error bars for the MDF distribution calculated? Showing the actual data points should be a clearer way to show what the raw distribution looks like.

Reply 22: Error bars were calculated in the TDMAinv software, and the procedure is described in detail by Gysel et al. (2009). The MDF is the measurement distribution function, and actually shows the raw distribution (corrected to 90% RH). i.e. the actual data points, for that scan.

Comment 23: Figures 2 and 4: Given the overlapping uncertainty envelopes, it is hard to distinguish which colour corresponds to which curve (especially at 165 nm). For example, red and magenta are nearly indistinguishable, as are purple and blue. Recasting these figures using the hygroscopicity distribution concept might be a clearer representation; otherwise, consider spinning each curve out into a separate subfigure.

Reply 23: These figures have been improved, and are also now complemented with kappa values as a function of diameter, similar to those of Su et al. 2010.

Comment 24: The stacked bar graphs in Figure 5-7, 10-11 are very interesting and a nice contribution to better quantitative understanding of the aerosol mixing state.

Reply 24: Thank you.

Minor comments:

Comment 25: Pg. 6760, Line 4: Research was conducted under this period but the data set is not continuous. This should be noted here as it is on Pg. 6767, Lines 17-21.

Reply 25: This has been clarified in the text: *"A hygroscopicity tandem differential mobility analyzer (HTDMA) was used to evaluate the hygroscopic properties of aerosol particles measured at the Puy de*

Dôme research station in central France, periodically from September 2008 to January 2010, and almost continuously from October 2010 to December 2012."

Comment 26: Pg. 6760, Lines 17-20: I don't think speculative conclusions are appropriate for an abstract.

Reply 26: Yes, the speculative phrasing has been removed.

Comment 27: Pg. 6761, Line 12: Update to the latest IPCC report.

Reply 27: Reference has been updated.

Comment 28: Pg. 6762, Lines 12-13: Boundary layer aerosols often feed convectively-driven tropospheric clouds.

Reply 28: Yes, this is now mentioned in the text: *"For the formation of convective clouds (e.g., cumulus), boundary layer (BL) aerosols also play an important role."*

Comment 29: Pg. 6762, Line 25: Again, please note that the data set is not continuous.

Reply 29: This has been clarified in the text. *"The work presented covers data collected in periods from September 2008 to January 2010, and almost continuously from October 2010 to December 2012. This is, to the authors' knowledge, the first study that investigates hygroscopic properties of particles measured at a high altitude site almost continuously over several years. This allows for detailed analysis of the influence of year-to-year and seasonal cycles, diurnal patterns, and air mass types on particle hygroscopicity."*

Comment 30: Pg. 6763, Line 18: Remove citation to Hervo et al., 2014, which is in preparation. It is grossly misleading and inappropriate to cite a non-published paper with a publication year.

Reply 30: The citation has been removed.

Comment 31: Pg. 6765, Line 1: Was the RH constant at 90% or was it varied. Why is recalculation necessary?

Reply 31: The measured RH value varies over the course of the measurements. In general this variation is small, ±3%, however in some cases the variation can be larger. The data analysis procedure filters out any data that was measured with a greater range of RH than 87 to 93% and corrects all the others to 90% so that the measured GF are comparable.

Comment 32: Pg. 6765, Lines 22-26: Is this referring to the GF and sigma for each mode or for the entire PDF?

Reply 32: This is referring to the entire PDF. A large sigma suggests that the PDF should be divided into different modes.

Comment 33: Pg. 6766, Line 5: Reference Figure 8 here.

Reply 33: Thanks, reference to Figure 8 is added.

Comment 34: Pg. 6766, Line 5: What is the "oceanic modified" air mass type referring to? How is the air mass modified?

Reply 34: Oceanic modified refers to air masses originating from the ocean, but have travelled over the continent and are thus influenced by continental sources. This has been clarified in the text: *"The classification oceanic modified refers to air masses originating from the ocean, which have travelled over the continent and are thus influenced by continental sources."*

Comment 35: Pg. 6770, Line 5: What is "modified sea salt"?

Reply 35: Modified sea salt refers to sea salt that has been modified by other species in the air, for example condensation of secondary continental species. This has been clarified in the text: *"Modified*

sea salt refers to sea salt that has been modified by other species in the air, for example condensation of secondary continental species."

Comment 36: Pg. 6772, Lines 23-24: Stating that the less hygroscopic fraction increases during the day at the expense of the hygroscopic fraction does not tell the reader whether there are more less hygroscopic particles present or just fewer hygroscopic particles present during the day.

Reply 36: The changes are, of course, for one population relative to the other. We have now stressed this in the text: "*This diurnal variation is due to the less hygroscopic fraction that increases during the day, while the hygroscopic fraction decreases."*

Comment 37: Pg. 6773, Line 19: What height was used to initialize the back trajectory?

Reply 37: The HYSPLIT air mass backward trajectories were calculated for the arrival pressure 850 hPa at the height of the puy de Dôme. This has been added to section 2.3.2 of the text. *"Trajectories were calculated for the arrival pressure 850 hPa at the height of the pdD every 6 hours (…) "*

Comment 38: Pg. 6775, Line 16: Does CNN refer to "cloud condensation nuclei (CCN)"?

Reply 38: Yes, thanks. This is corrected.

Comment 39: Pg. 6778, Line 27: I don't think "realistic" is the correct word here, as ambient RH conditions can vary from dry air masses to moist air masses.

Reply 39: Thanks, we have changed the word to "useful".

Comment 40: Pg. 6780, Line 29: Strike the Hervo et al., in preparation reference since it is not yet accepted for publication.

Reply 40: The reference has been removed.

Comment 43: Table 1 could be moved to the supplementary material as it is not directly relevant to the discussion.

Reply 43: Yes, the table has been moved to the supplementary material.

Comment 44: In Tables 2-4 and Figures 1-7, 10-11, please explicitly note that the GF is GF(90%) in either the caption or the Table heading/Figure axis labels.

Reply 44: Thanks, this has been added to the captions.

References

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