Referee 2: we thank Referee 2 for his comments.

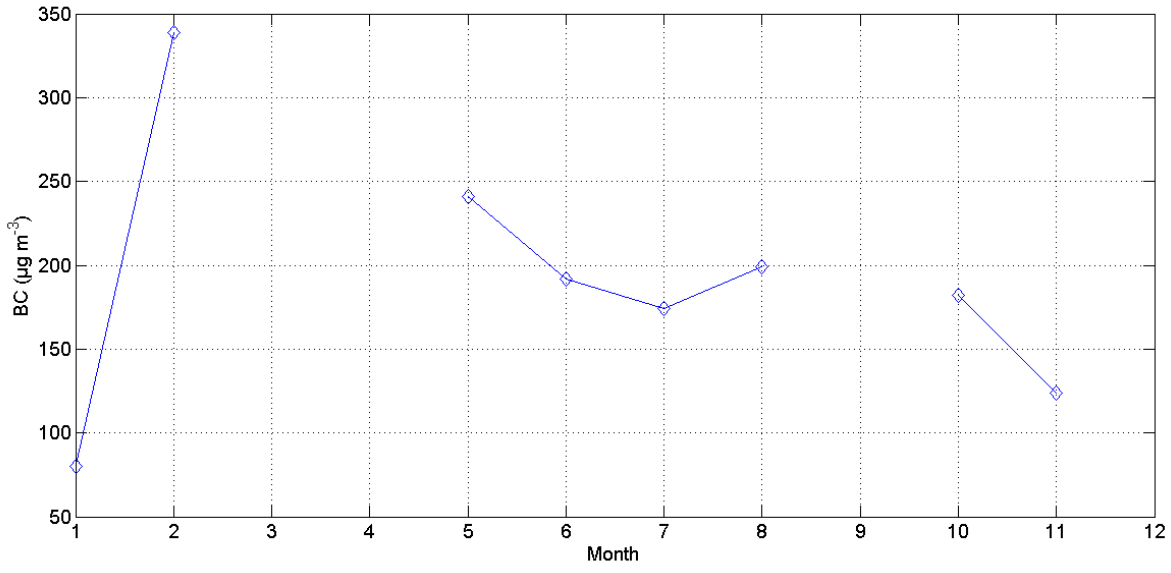
Comment 1: Major comment: Discussion Paper Already in Quick Access Review I have asked authors to elaborate more on attributing C6813 the NPF to free troposphere. This comment was not addressed at all. Oneadditional ACPD sentence referring to [Boulon et al., 2011]. Boulon et al investigated use of lidar in estimation of planetary boundary layer. On page 5627 they compare WTC method 14, C6813–C6816, 2014 used in this manuscript with their own method derived for conditions in a vicinity of Puy de Dome: The method proposed here was compared to the WCT algorithm proposed by Brooks (2003), initially developed for marine boundary layer height retrievals. The Interactive calculated PBL height was found to be 32.2 % higher on average when it was com- Comment puted using our method. This difference comes from the fact that the WCT method tries to find the upper limit of the most concentrated aerosol layer (i.e. the start of the decrease of the Mie regime) whereas our method was build to find the transition from Mie diffusion regime to Rayleigh diffusion regime i.e. the transition from planetary boundary layer influenced layers to free tropospheric influenced layers. Compared to WCT and according to LIDAR contour plot, this procedure seems to be better adapted to the calculation of the PBL height in mountainous area such as Puy de Dome. I won- der why authors did not use the more appropriate method from this article instead of WCT method. Adding approximately 30% to WCT method to data presented in Fig- ure 1, the PBL is at or above altitude of Puy de Dome and we cannot any more talk about nucleation in free troposphere, but in upper part of PBL. This is also supported by Figure 4. Claiming that black carbon concentrations > 1 μg*m-3 are relevant for free troposphere does not find any support in literature. Moreover, during the NPF periods 1 and 3, the synoptic situation was controlled by strong high pressure system. During Full Screen / Esc these conditions free troposphere is dominated by large scale subsidence and there- fore very low relative humidity in lower FT. Measurements show opposite, high humidity Printer-friendly Version between 90 – 100% for period 1 indicates that station itself was most likely directly in- fluenced by orographic upslope flow of boundary layer air from valley below. Sharp rise of RH during period 3 on 28 Feb indicates the same process. This is also in agree- ment with absolute values of condensational sink and BC for period 1. For Period 3 Discussion Paper some additional tracer is needed due to absence of BC measurements. This behavior is typical for all mountain stations with late morning- noon rise of nearby BL upslope to C6814 the mountain top depending on local orography and altitude difference between moun- ACPD tain top and surrounding areas. The rise of the air mass associated with decrease in pressure and temperature by itself create conditions more suitable for NPF. Authors in- 14, C6813–C6816, 2014 terpret data as free tropospheric feeding process with impression that it is a large scale feature, but without discussing also altitude evolution of the air mass trajectories dur- ing transport it is only unsupported speculation. Again, they have to show much more Interactive rigorous and complete analysis to distinguish between free troposphere and boundary Comment layer. There are many more parameters available at Puy de Dome. I would like to see corresponding diurnal cycle

of total aerosol number density, size distribution, trace gases and meteorology, especially local wind and its variability.

Reply 1: Concerning the WCT method, it is true that it was initially developed in the marine atmosphere but it has already been used in different environments since (eg: Baars et al., 2008, Leipzig, Germany). In their study, Boulon et al. (2011) developed an alternative method which was reported to give BL heights 32% higher on average compared to the WCT method. When using this procedure with our dataset, we find BL heights that are on average only 8% higher compared to those derived from the WCT (see Reply 3). Thus it seems that the determination of BL/FT from LIDAR measurements is not impacted by the method that we use.

However, as suggested by Referee 2, we have added to the manuscript time series of atmospheric parameters that can help to distinguish between the BL and the FT (SMPS size distribution, NO_2 concentration and wind speed) and which lead us to slightly nuance our analysis. In fact, during Period 1, the presence of an accumulation mode on the SMPS size distribution, high NO_2 concentrations and high variability in the wind speed measurements lead us to conclude that the station was at the BL/FT interface rather than in the FT in the morning, especially on the 10th and 11th. During Periods 2 and 3, these additional parameters confirm LIDAR measurements which suggest that the station is sometimes in the BL, but always out of the nucleation hours, during which the station is in the FT. Thus, during nucleation hours, Period 1 is now discussed as representative of interface conditions between the BL and the FT and Periods 2 and 3 are still considered representative of FT conditions. Changes were provided throughout section 3 according to this re-analysis.

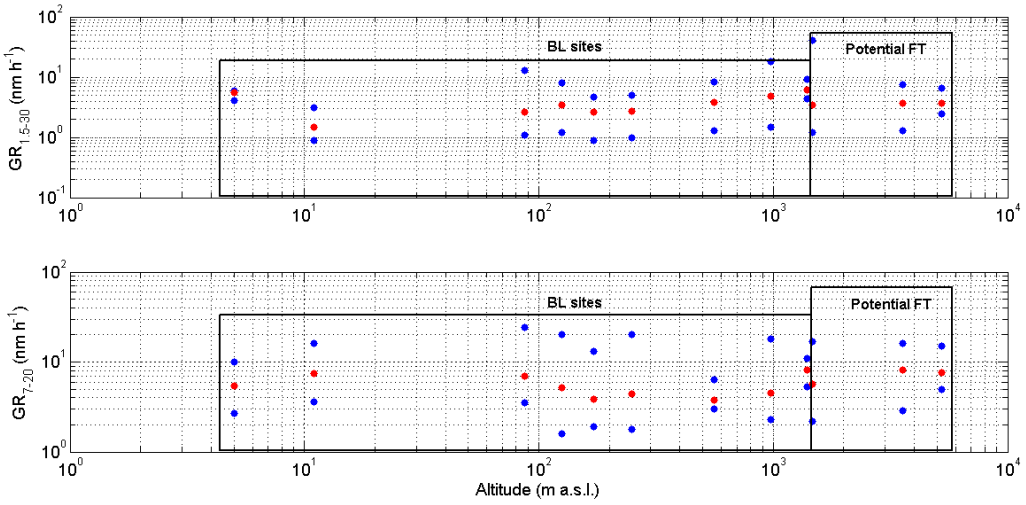
In the description of the sub-periods, CO concentrations were added to supply the lack of BC data during Period 3 and show variations which are in agreement with BC and CS. Concerning BC concentrations, we do not claim that they are relevant for free troposphere conditions, we only notice that they are relevant for the station. This is clearly visible on the figure below, which shows the monthly mean BC concentrations for 2012.



At last, hypothesis on a “free troposphere feeding process” was removed, and the influence of long range transport from Eastern Europe was only mentioned as a potential additional source of pollution.

Comment 2: Abstract L3-4: With respect to slow growth into CCN size in FT compared to BL I do not understand what authors mean by special importance of direct influence on cloud formation.

Reply 2: The authors mean that the number of CN and hence CCN are particularly relevant at high altitude where clouds form. Referee 2 asserts that particle growth is slower in FT compared to BL. It might be true, even if it is not obvious that particle growth decreases with altitude, as shown on the figure below:

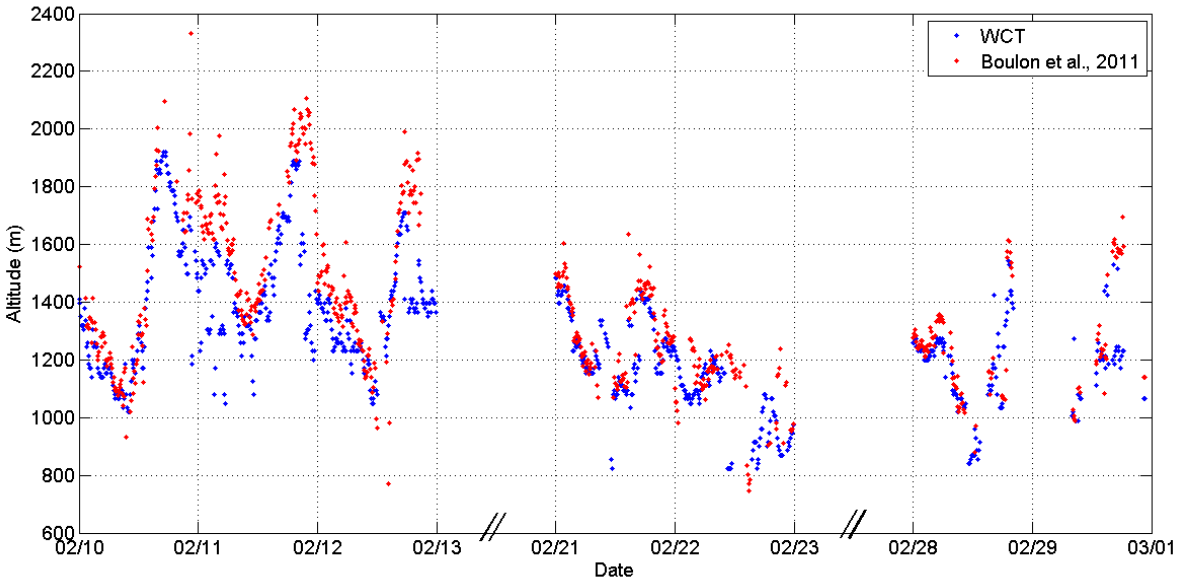


This figure shows the ranges of GRs (blue) as well as the average values, mean or median (red), from different sites as a function of the altitude of the station (Manninen et al., 2010; Vakkari et al., 2011; Rose et al., in prep) for the size range 1.5 – 3 nm (upper panel) and 7 – 20 nm (lower panel). We can see that for the highest stations, which can periodically lay in the FT (i.e. Puy de Dôme, Jungfraujoch and Chacaltaya (5240 m, Bolivia)), the GRs are not significantly lower than the GRs observed for BL stations.

In any case, regardless of the variations of the GR with altitude and the comparisons between the BL and the FT, model studies such as Merikanto et al. (2009) clearly report that NPF occurring in the FT is a significant CCN source for the whole atmospheric column. Especially, it is reported that “The model suggests that 35% of CCN (0.2%) in global low-level clouds were created in the free and upper troposphere”.

Comment 3: P18264 L8-10: In opposite, Boulon et al states that WTC is not suitable method for Puy de Dome and their new method delivers better results in estimation of PBL using lidar signal (see part with major comment). Authors here create false impression that they have chosen correct algorithm to estimate PBL from lidar signal.

Reply 3: In their study, Boulon et al. (2011) say that their method "seems to be better adapted", but they do not give clear reasons to reject the WCT procedure. We have compared the BL heights derived from the two methods for the period of interest. The result is shown on the figure below.



The two procedures give very similar results: the method from Boulon et al. (2011) gives results that are only 8% higher on average compared to the WCT, which is significantly lower than the 32% reported by Boulon et al. (2011) and does not drastically change our conclusions. Thus we do assume that the WCT, widely accepted as one of the most reliable method in the literature, can be considered as a correct algorithm for the present study.

Comment 4: P 18367: Did authors try to use ratio between PSM and ions instead of introducing arbitrary constant of 500 cm-3?

Reply 4: As mentioned in the manuscript, recent studies such as the one by Wimmer et al. (2013) suggest that the PSM could be unable to detect all cluster ions because of their chemical composition. However, we do not know if the PSM does not detect a constant fraction of the charged clusters, of if this fraction is proportional to the charged cluster concentration. Thus, in order to be as cautious as possible and obtain a lower estimation of

the actual total cluster concentration, we chose to use a lower detection limit, which might be considered as arbitrary but was in fact derived from the measurements.

Comment 5: P18368: here should be also included study of the charged particles distribution in European troposphere from EUCAARI experiment [Mirme et al., 2010]. It provides good information about the background FT concentrations and vertical distribution.

Reply 5: the reference was added. However, altitude scales and especially diameter ranges (2.5 – 3 nm for charged and total clusters, and 0.75 - 2 nm for charged clusters)used in Mirme et al. (2010) do not allow a direct quantitative comparison. This is the reason why only major qualitative results were included in the manuscript as follow: "…with lower values at night. NAIS airborne measurements from the whole tropospheric column (up to 12 km) conducted in the frame of the EUCAARI – LONGREX campaign (May 2008) were reported by Mirme et al. (2010) and showed similar results. In fact, charged clusters were continuously detected at all altitudes with a mode centred around 1 nm. Between 2.5 and 3 nm, total cluster concentrations significantly exceeded charged concentrations, suggesting a continuous pool of sub -3nm neutral clusters throughout the whole tropospheric column.

The first observations…"

Comment 6: Figure 4: One needs microscope to see properly what is included. The figure needs improvement in size and scales.

Reply 6: We have tried to improve the quality of Fig. 4. CO concentrations were added to supply the lack of BC measurement and the location of the station in the BL/FT was also indicated.

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