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Interactive comment on "Major contribution of neutral clusters to new particle formation in the free troposphere" *by* C. Rose et al.

Anonymous Referee #2

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The paper "Major contribution of neutral clusters to new particle formation in the free troposphere" by Rose et. al. presents observations of the new particle formation (NPF) events including neutral and charged clusters observed during a short period in February 2012 at Puy de Dome mountain station in France. The results are interpreted with help of limited set of meteorological parameters, lidar observations, black carbon and air mass back trajectory analysis. The main conclusions of the article is 1) importance of neutral clusters in NPF 2) interpreting the observations as NPF occurrence in free troposphere, 3) NPF is nor sulfuric acid limited.

Major comment:

Already in Quick Access Review I have asked authors to elaborate more on attributing the NPF to free troposphere. This comment was not addressed at all. One additional 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 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 calculated PBL height was found to be 32.2 % higher on average when it was computed 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 wonder 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 Figure 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 these conditions free troposphere is dominated by large scale subsidence and therefore very low relative humidity in lower FT. Measurements show opposite, high humidity between 90 – 100% for period 1 indicates that station itself was most likely directly influenced 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 agreement with absolute values of condensational sink and BC for period 1. For Period 3 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 the mountain top depending on local orography and altitude difference between mountain 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 interpret 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 during transport it is only unsupported speculation. Again, they have to show much more rigorous and complete analysis to distinguish between free troposphere and boundary 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.

Detail comments:

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

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.

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

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.

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

At current level of data analysis and interpretation I do not believe that NPF can be at-

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Interactive Comment

tributed to FT conditions and authors cannot support their claims of observing NPF in FT conditions. Than the main novelty of the article is not supported by rigorous analysis and interpretation. Use of inappropriate lidar algorithm only supports that. Available information points more towards influence of local upslope wind of Puy de Dome and not a free tropospheric feeding process of larger spatial importance as claimed by authors. Statement about observing NPF in free troposphere is not sufficiently supported by analysis presented in the manuscript. In current form I cannot recommend the manuscript for publication in ACP and major revision is necessary.

References

Boulon, J., K. Sellegri, M. Hervo, D. Picard, J. M. Pichon, P. Freville, and P. Laj (2011), Investigation of nucleation events vertical extent: a long term study at two different altitude sites, Atmos. Chem. Phys., 11(12), 5625-5639. Mirme, S., A. Mirme, A. Minikin, A. Petzold, U. Horrak, V. M. Kerminen, and M. Kulmala (2010), Atmospheric sub-3 nm particles at high altitudes, Atmos. Chem. Phys., 10(2), 437-451.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 18355, 2014.