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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Seasonal variation of aerosol size distribution at Puy de Dôme (1465 m a.s.l., central France)" *by* H. Venzac et al.

H. Venzac et al.

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Referee Number 1 We fully agree with the referee’s general comment and we have now better emphasis the fact that we do not describe only the FT composition but also possibly the residual layer composition, even though we applied an additional filter to our “free tropospheric data”, based on the modelled BLH and calculation of splitting height Hs. The “Free Troposphere” term is changed to “Free troposphere/residual” through the paper.

Specific comments Regarding terminology and English expressions the manuscript needs improvements. I just give some examples here: "aerosol concentrations" should be more specific: "aerosol number concentrations"; "air mass concentrations" (and similar phrases) do not exist, it should read "particle number concentrations... in particular air masses"; "ECMWF" should be "ECMWF data or products"; modes of size

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distributions should be adequately described etc. We improved the manuscript regarding the terminology.

Page 15794, line 10. Nucleation mode definition: I would say the nucleation mode extends down to few nm rather than 10 nm as stated. It is only in this study that there are no measurement for particles smaller than 10 nm available. A comment on the fact that particles below 10 nm could not be measured in this study is needed later on in the text. Yes, we modified the text in the following way :

“Current knowledge of aerosols number size distributions has generally identified the number size distribution to be composed of a Nucleation mode (3-20 nm) representative of recent new particle formation, ..” and later in the experimental part: “The process of nucleation itself can not be examined in the present study, but some information can be extracted on the occurrence of new ultrafine particles formed by secondary processes, as 10-nm particles are of primary origin only when sampled very close to their source. ”

Page 15794, line 16. Why exactly 36 hours? This is an typing mistake, in fact the air mass back trajectory calculations are performed on 72 hours, and we modified the text accordingly. We arbitrarily chose 72 hours because the turnover time of aerosol particles has been evaluated to be from 1.6-1.7 days for nuclei size ranges, to 2.4 days for 200 nm particles (Tunved et al. 2005). This is now specified in the text. Section 2, page 15795/6. Do I understand correctly, that the data set used later on includes more than 50 % of in-cloud data? If not, it should be clearly stated. If yes, the authors need to discuss this (later in the manuscript). It is not clear at all if in-cloud and out-of-cloud measurements of aerosol size distribution can be combined into one meaningful data set. In-cloud data are included, but, as stated in the text: “Because the Puy de Dˆome station is more than 50% of the time in-cloud, the aerosol sampling is performed through a whole air inlet (WAI) which ensures efficient sampling of both cloud droplets and interstitial aerosol in the presence of clouds. Based upon theoretical considerations, the WAI is capable of efficiently sampling droplets $<$35 μm

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for wind-speed <10ms−1. During cloudy conditions, interstitial aerosols and evaporated cloud residues are sampled simultaneously at a relative humidity close to 50%.” This means that we artificially evaporate the cloud in the WAI, and we should find the same size distribution than immediately before the cloud formed on the site, and immediately after the cloud disappeared (except if cloud processing of the aerosol in cloud is significant). This method is commonly used at thigh altitude stations to exclude the sampling artefacts due to the presence of clouds. It is now better explained: “Because the Puy de Dôme station is more than 50% of the time in-cloud, the aerosol sampling is performed through a whole air inlet (WAI) which samples during cloudy conditions both aerosols and cloud droplets which are subsequently evaporated to cloud residues (Sellegri et al. 2003). Based upon theoretical considerations, the WAI is capable of efficiently sampling droplets <35 μm for wind-speed <10ms−1, which represents most of the cloud droplets population in a typical cloud. The resulting aerosol size distribution is equivalent to the one which would be obtained after a natural dissipation of cloud”

Page 15796, lines 12 and following. State briefly how the inversion of the SMPS data was done for this instrument. We briefly described the main characteristics of our inversion.

Section 3.1, page 15798, lines 14-17. Why this reference to horizontal advection of air with variable BL height? Isn’t the more direct and simple explanation the growth of the BL over the height of the mountain top? Indeed, the sentence has been changed to: “The increase of concentrations during the day observed at the puy de Dôme station can result from nucleation events (Venzac et al., 2007), but also be resulting from the growth of the boundary layer over the height of the mountain top.”

Section 3.2, page 15798, lines 19.. Size distribution data of the SMPS are available for 2006 only. 2006 is the year with the lowest overall concentrations in the long-term data set. Can the authors comment on that? In the absence of modelling studies, it is

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difficult to identify causes for lower CN concentrations in 2006. In fact, it appears that the decrease is due to low concentrations during summer 2006 while winter concentrations are similar to those in previous or successive years. First, we can exclude an instrumental artefact due to the fact that the total concentrations issued from the SMPS might underestimate the concentrations measured with CPC. Indeed, not only winter time concentrations are similar but during 2007 during which we report only CPC total counting concentrations are quite similar to those of 2006. As reported in the method section, the comparison between SMPS total counting and CPC is of the order of 5% for ambient aerosol during EUSAAR intercomparison workshops. The suggestion to link low concentration values to meteorological parameters is certainly of interest. In fact 2006 was relatively cold compared to the previous years and this might have influenced the summer max concentrations. It is now commented in the text. However, because the link between T at PDD and CN concentration is not a direct cause to effect relationship, we do not feel adding a new figure which may be subject to criticisms.

Section 3.2 in general. I would prefer in 3.2 the same structure as in Section 3.1: First discuss long-term and seasonal variations, then discuss diurnal variations. Check in particular 2nd and 3rd sentence of this section. I find this confusing. We arranged the structure of section 3.2 according to the referee’s suggestions

Page 15798, lines 23-24. "modes around ..." What exactly do the authors refer to? Maximum of the mode? But apart from that, I cannot see well the modes in Figure 3. The seasonal evolution of the modal diameters are commented in detail later in the section, hence we chose to remove this sentence.

Page 15798, last line and following lines next page. I am not sure I understand this sentence. Clearly, the authors should explain in more detail how often nucleation events were observed and how this shows up in the size distributions they discuss. This question has also been raised by the referee #3 and we agree that nucleation should be partly described here. The text is now: “The nucleation mode that we describe in the present paper is in fact an already grown nucleation mode, since we detect it

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after particles have reached diameters larger than 10 nm. An adequate instrument (Air Ion Spectrometer, AIS) has been used to track nucleation events down to the sub-nanometer size, and show that nucleation is observed on average more than half of the measurement days, with a minimum frequency of 29 % during winter (Venzac et al. 2007, and unpublished data). The solar radiation reaching the puy de Dôme station shows a significantly higher median value for nucleation event days than for non event days, indicating that nucleation seems favoured under clear sky conditions. A full description of the seasonal variation of nucleation and new particle formation events will be described in a separate paper. New particle formation and growth is defined by the occurrence of higher concentrations of 10 nm particles at mid-day and their following growth of 10 nm particles to larger sizes (up to the aitken mode size). Due to their relatively high frequency, the impact of new particle formation and growth events is detectable on the median size distribution shown Figure 3 for all seasons. The median growth rate of 10 nm particles which can be inferred from Figure 3 are 5.6, 5.2, 4.8 and 4.7 nm h-1 respectively for spring, summer, autumn and winter.”

Page 15799, lines 3-7. Some explanation is needed how the fitting of the 3 lognormal modes was actually done and how well it represents the measured size distribution. In reference to the previous comment and what is shown in Figure 4, I don’t see "the impact of nucleation events". It is difficult to characterise the nucleation mode if the measurement does not extend to below 10 nm particle size. We agree that nucleation has not been measured here, and only “new particle formation events” can be detected from the 10 nm size. We now use the appropriate term. The impact of new particle formation and growth is clearly visible with the occurrence of higher concentrations of 10 nm particles at mid-day and their following growth of 10 nm particles to larger sizes (up to the aitken mode size) on Figure 3, especially during summer. It is now better explained in the text. Regarding the fitting, the fitting procedure is now better described. It is indeed difficult to fit the nucleation mode when it is only partly detected. Neither the size nor the concentration is well captured by the fitting procedure when the 10 nm particles burst appear in the SMPS size distribution. We did not find

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any solution for this problem, and consider that the fitting is satisfactory only when the particles have reached about 20-25 nm. This aspect is now better explained in the text. Our observations can be used for comparing relative size and concentrations between seasons. The nucleation mode is of course not complete quantitatively, and we now clarify this. The impact of nucleation events can be seen in Figure 4 by the presence of a nucleation mode on average size distributions during the day in comparison to the nighttime average size distribution.

Page 15800 lines 10-13. "Higher influence" on what? In general, I think the influence of the BL height as one of the largest driving factors for aerosol variability at a mountain location should be mentioned already in Section 3.1 (and discussed in detail in Section 4). The BL height is mentioned section 3.1. as a driving factor for the aerosol variability and the line 10-13 p 15800 modified according to the referee comment.

Section 4.1. A number of points need to be addressed here: Did the authors use analysis data of the ECMWF model? How was the BL height determined? Can one show the daily variation of the BL height for different seasons? (See also my general comment.) We do show the daily variation of the BL height calculated from the ECMWF model for different season. We do not understand the referee’s request.

Are there any meteorological or other in situ data sampled at the Puy de Dome observation site which support this analysis based on ECMWF data? This question has been raised by referee #3 as well. It is very difficult to find a parameter which determines if the puy de Dôme station is within the BL or not. Rnd, which would be the parameter which would be the closest to give this information, can not be used alone, as it also varies between continental and marine air masses. The puy de Dôme is now equipped (since very recently) with a LIDAR which will provide BL heights, but this data was not available at the time.

Page 15801, lines 12-14. I doubt this is the case (see general comment) and the authors need to more carefully argue here. We took the referee’s general com-

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ment and specific remark in to account and we are now more careful: “Results confirm that the air reaching the puy de Dôme sampling site is strongly influenced by the BL during the day during summer, while, on average, the model shows that the site lies above the boundary layer, either in the residual layer or in the free troposphere (FT) during winter. At night, the modeled BL height is lower than 500 m a.s.l. for all seasons.”

A reference is needed for the discussion of Froude number etc. (e.g. Baines) Done

Page 15802, lines 5-6. Which two regimes are the authors referring to in this sentence? How was Hs calculated? Were the values for virtual potential temperature (gradient) and wind speed taken from ECMWF data or from actual measurements? Our text was not clear and we modified it: “For stable conditions, a splitting height Hs can be calculated according to Eq. (3): Hs=H(1-F) (3) For an altitude lower than Hs, the flux will deviate from the obstacle while at altitudes higher than Hs it will be uplifted along the mountain slope.”

Page 15802, lines 16-17. What are vertical trajectories? This needs to rephrased. This sentence was rephrased.

Page 15802, lines 23-24. This refers to Figures 4 and 5, apparently. This should be stated. Apart from the fact that I doubt the approach to just focus on night time data to obtain a meaningful data set representative for the FT, the long-term record (like in Figure 1) should be shown explicitly for night-time conditions, if the authors want to follow up on this. We believe that the seasonal variation of nighttime concentrations are clearly shown on figure 3 (previously figure 4), but we followed the referee’s suggestion to add a figure showing the long-term record of nightime data (Figure 1b).

Page 15803, line 4. Why "at this stage"? This was rephrased

Page 15804, lines 18-25. Very inaccurate phrasing here! The differences observed can only refer to concentration properties. An air mass has no concentration. This was

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rephrased: “On average, at night and in marine air masses, aerosol particles are 4.4ś1.8 times more concentrated during summer than during winter for a same transport route. In continental air masses, we also observe an increase of the aerosol particle concentrations by a factor of 5.8ś1.3 from winter to summer.”

Page 15805, line 4. "Total aerosol column load"? What is this? I have the feeling that the discussion in general is mainly based on total number concentrations in the SMPS size range. This total concentration is usually dominated by the smaller particles in the size range. The authors should consider discussing the accumulation mode integral number concentration separately from the total concentrations. By aerosol column load, we meant total aerosol concentrations integrated over the whole atmospheric column. It is now clarified in the text. We also better discussed the contribution of the nucleation, aitken and accumulation modes to the total aerosol concentration increase from winter to summer.

Page 15805, lines 15-17. The definition for the longitude ranges appear to be mixed up here. Yes we corrected this.

Page 15806, lines 18-23. This is a comparison between marine BL air and allegedly FT air. In how far does this make sense? It makes sense if the aerosol sampled at the PDD have been transported at high altitude after they have been vertically transported above the MBL. Dust has frequently been observed at PDD, sea salt as well. We believe BL air from sahara or the Atlantic ocean should be compared to their concentrations far away from their source region: how are they ageing?

Figure 1. Include also a night time concentration only graph, if this is still considered to be more representative of FT conditions. Figure 3. Why is the diameter range different in the four panels? Data outside SMPS size range should not appear. Figure 5. Add in the caption that these are measurements of 2006. Are the data robust against the choice of day time / night time definitions? Figure 1 now includes night time concentrations only. Figure 3 has been changed to Figure 4, the range of diameters

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has been homogenized Figure 5 has been replaced by Table 1, as requested by referee nb 3.

Figure 8. The figure caption is very misleading. These are no maps of aerosol number concentrations! Rather this indicates the relationship between observed concentrations (at a single site) and air mass origin (based on a 3 day transport time). This needs to be carefully explained. In general, I wonder if the resolution is not too crude to resolve dependencies of aerosol concentrations on transport patterns. How did the authors attribute for the altitude history of the air transported to the observation site? We modified Figures 7 and 8 captions, and their descriptions in the text. The season-segregated maps sometimes contain areas where only one or two air mass trajectories end: increasing the spatial resolution would decrease too much the statistics on such plots, especially for Figure 8 for which average concentrations are calculated. We did not consider the altitude history on Figures 7 and 8, but studied it separately (figure not shown) in order to check for a seasonal variability. The combination between geographical origin and the altitude history in the same graph would be very valuable, indeed; but we do not possess the adequate tools for such a representation right now.

TECHNICAL COMMENTS Introduction, page 15793, line 22. "Dp": "p" should be written as index. Pages 15795, line 6. "sites" (s missing) Page 15806, lines 1-2. The characteristic parameters are not shown in Figure 9, only in the table. Corrected

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Interactive comment on Atmos. Chem. Phys. Discuss., 8, 15791, 2008.