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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.

Anonymous Referee #3

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General comments

The authors report on atmospheric particle number concentrations and size distributions collected on top of the Puy de Dôme, a solitary mountain in the French Massif Central. Number concentrations were presented for a duration of six years, while size distributions are discussed for a one year period. The authors present median values of their data, and discuss the measurements as a function of season, time of day, and air mass origin.

The paper is a valuable contribution to the growing literature on atmospheric aerosol parameters observed around the globe. In general, I like the way the authors present their material, graphs etc.

Meanwhile, the results obtained by the authors are not spectacular, and I am a bit disappointed that the the data analyses were not pursued more persistently. To meet the high standards set by other ACP publications, improvements are clearly necessary involving additional data analysis. I therefore recommend a publication in ACP only after the below mentioned issues have been addressed.

My main observation is that the paper remains on a quite descriptive level. The authors provide statistical values of their data, but the really new conclusions about the atmospheric processes leading to these values remain superficial. The authors report size distributions encompassing one year. While it is acknowledgeable that they introduce data from a new observation site to the public, similar studies on the same topic have been around for several years.

The authors also argue that their night-time data be representative for the free troposphere all around the year which is, in my view, only vaguely supported (specific point 8c) below). As a whole, I recommend a more thorough analysis of the data, under the inclusion of additional atmospheric parameters. I have also various rather technical comments, included in the detailed list below.

Abstract and Introduction

1) Particle number concentration and size distributions are mentioned as key parameters to constrain uncertainties in the prediction of atmospheric radiative effects. The radiative effects, however, are essentially related to light scattering, absorption, and CCN concentration. These are quantities that are different from those measured. It needs to be outlined in a few sentences how light scattering, absorption, and CCN concentration are related to, or could be derived from the measured magnitudes.

2) There is a need to mention the size range of the SMPS measurements as well as the effective cut-off size of the CPC (including the inlet system) in the abstract and the conclusions.

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Sampling techniques and site

3) The authors write that “Meteorological parameters, including the wind speed and direction, temperature, pressure, relative humidity and radiation (global, UV and diffuse), atmospheric trace gases (O3, NOx, SO2, CO2) and particulate equivalent black carbon (eqBC) are monitored continuously throughout the year.” Unfortunately, most of these data were not referred to any more in the paper. Also, http://wwwobs.univ-bpclermont.fr reports that concentrations of Radon are collected at Puy de Dôme, as well as size distributions of particles up to 10 μm with an optical particle counter.

I am wondering why none of the many additional data sources available on-site have been tapped to substantiate the conclusions made in the paper. The paper would be significantly enhanced if the authors consider relating their aerosol measurements to those key trace gas and meteorological parameters.

It would be valuable to see a statistical analysis, such as a factor or principal component analysis of all the measured parameters. That might help to examine how smaller and bigger particles correlate, how they relate to solar radiation, trace gases etc., and help distinguish the different regimes boundary layer/free troposphere.

4) There are several technical questions regarding the collection of the number and size distribution data.

4a) There is no mention on the relative humidity neither inside the DMA, nor the charge neutralizer. Were they controlled and/or measured? They can have a great effect on the sizing of the particles as well as the correct determination of the charging efficiency. These issues are vital for the comparability towards other data sets.

4b) “Inter-calibration within the EUSAAR project”: The particle size during this inter-calibration deviated “20% for the 20 nm particles and 10% for the 100 nm particles”. This is quite a lot for differential mobility analyzers. A deviation of 10% in size propagates non-linearly into the calculation, e.g. of scattering coefficients. There is also

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no mention whether the deviation was upwards or downwards. What kind of aerosol was used during the inter-calibration? Can the deviations in particle size be explained somehow?

4c) Also, the SMPS was operated at a scanning time of 2 min. This relatively short scanning duration might lead to a smearing of the measured size distribution (Collins et al., Aerosol Science and Technology, 38:833–850, 2004). This might also be connected to the mentioned deviation in particle sizing. Could the authors, e.g. on the basis of some test measurements using monodisperse particles, or a comparison of upward and downward scans, confirm that such a relatively short scanning time had no smearing and distorting effect on the size distribution?

4d) I did not find any documentation of the particle losses, e.g. by diffusion, in the inlet lines. This, however, could have a great effect on the measured size distributions. Especially the CPC measurements could be degraded in the case of particle losses above 10 nm. Were the size distributions corrected for any losses in the inlet lines? (CPC data cannot be corrected for inlet losses since the actual size of the particles is not known.) Again, these issues are vital for the comparability of the data towards other data sets, which I assume is the intention of the authors as well.

Seasonal and diurnal variability...

The title of this section is very long.

Particle number

5a) Besides the seasonal variation in particle number, quite a significant inter-annual variability can be observed (Fig. 1). 2005 shows a quite anomalous annual pattern. 2006 and 2007 show considerably lower numbers than the years before. At the same time, the minimum values in Fig. 1 appear to show a less pronounced decrease over the years. The decreasing trend seems to be mainly attached to the higher values occurring in summer. It would be worth to explore the reasons for these features. Can

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they be explained in meteorological terms (temperature, sunshine, winds)? If particle number in summer stems from the boundary layer, was there a similar trend in anthropogenic trace gases (CO) and radon? Was maybe the mixed layer height different in 2006 and 2007 compared to the years before? Or could there be instrumental reasons?

5b) In my opinion, the authors should point out the limitations of the total number data, since its connection with specific particle modes remains blurred. The size distribution data are clearly more valuable since they allow such conclusions. To improve the value of the total number data it would be worth to state the particle diameters in the size distribution that correlate most with total particle number. Then a reader could have a clearer idea which particle size range determines the number at different times of day and year.

Size distributions

6a) p. 15798, l. 24 “background signal”. Please explain what you mean with “background signal”. In fact, not only ultrafine particles are added during the day, but also particles at bigger sizes (Fig. 3). Were the concentrations of ultrafine and accumulation mode particles always coupled on the diurnal scale? A factor analysis or correlation analysis could help here.

6b) Figure 3: Try to narrow down the display of the data to the size range actually measured. There are large blue areas which are actually void of data. The number concentration in Fig. 3 is in fact redundant, since comparable curves are shown in Fig. 2. Fig. 3 could be enhanced by showing the tracks of the lognormal mode diameters. (Modal diameters are, in fact, discussed in the text.) From the nucleation mode diameter, one would also get an impression of the particle nucleation mode growth rate. The latter aspect has not been addressed elsewhere in the paper, but is vital in view of a comparison of this data set to others, even if the authors plan to publish such an evaluation in more depth in another paper.

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6c) p. 15799, l. 19 “this is consistent with more nucleation events...”: The authors mention nucleation events, which seem so important for the diurnal pattern of particle number concentration. However, they supply very limited information on the frequency of these events, the particle growth rates, their relationship with solar radiation, etc. I feel that it is necessary in this paper to include these information at least in a compact form. This would help to attribute the mid-day total number concentration towards particle formation events, as well as other aerosol types.

6d) Figure 4: Some of the size distribution data range between 3 and 1000 nm, others between 10 and 400 nm. Surprisingly enough, the 3–1000 nm feature appears only in the night-time data. How is this possible?

6e) Figure 5 is not very appealing. It would be better replaced by a Table similar to Table 1.

Links between aerosol physical...

7a) This chapter is very heterogeneous. Sect. 4.1 and 4.2 discuss meteorology-only results. Sect. 4.3 shows results of a sector analysis of the aerosol data, which is not linked to the two former chapters. Sect. 4.3 would belong, more logically, to the chapter 5 or even 3, which are the main results chapter. I urge the authors to reconsider their organization of the chapters 3-5.

7b) Figure 6 shows the computed boundary layer heights. This is an impressive effort. Unfortunately, it seems that these results are only used qualitatively to explain the diurnal and seasonal cycles, and not used in conjunction with the aerosol data. It would be very interesting to see, for instance, whether the mid-day particle concentrations and size distributions showed any positive or negative correlation with the boundary layer height. This could be a central new result of the study presented.

7c) Figure 7 and 8: As much as I like the author’s data display on globes, I have difficulty in recognizing meaningful features. The graphs could concentrate on a much

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smaller area, say the North Atlantic and Europe. Meanwhile, the spatial resolution (only 10 degrees) should be enhanced so that structures of potential source regions within Europe gain shape.

7d) The description of the method yielding Figure 7 and 8 is not very clear. Other researchers have performed similar allocations of measured concentrations to areas. The authors could definitely refer to these methods, and/or pick up some additional ideas. (Charron et al., Atmos. Env., 35:1387-1393, 2001; Tscherwenka et al., Atmos. Env., 32:3941-3952, 1998; Charron et al., J. Geophys. Res., 113, D07202, 2008).

7e) I noticed that the authors did not consider vertical trends in their calculated trajectories, but suppose that these could have a substantial influence on particle number concentrations and size distributions (subsiding air masses from the free troposphere vs. boundary layer trajectories). Again, radon and trace gases could help to classify the data.

7f) A problem with Figure 8 is that it might be biased by the life-time of aerosol particles. The correct allocation of measured concentrations to geographical regions as presented in the Figure is only meaningful if the measured substance is inert, i.e. does not react or change inside the atmosphere. The authors need to include a discussion on the presumed life-time of the measured particle concentration in the atmosphere. This might in fact be variable depending on the weather conditions from different wind directions.

7g) Why was Figure 8 not produced for different particle modes using the one-year size distribution data. This would be more meaningful than the given result for total particle number.

7h) Figure 9: Some of the size distributions fall down to zero above 200 nm. How is this possible?

7i) It would be interesting to learn whether one can see any influence of Clermont-

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Ferrand, the nearby city of 150,000 inhabitants, in the data. A city so close could artificially increase the reported number concentrations and size distributions values, especially for the continental air mass type.

Summary...

8a) p. 15809, l. 18 “Frequent episodes of new particle formation”. How many events, what growth rates (see above)? There is no need to provide a lengthy list, but a few numbers that can be compared to other data sets.

8b) Keep in mind that the size distributions from Whitby (1978) and Jaenicke (1993) were collected with equipment much more basic than the instrumentation used today. Most of their measurement campaigns lasted only relatively shortly. The data series collected since about 1995 are much more representative. It is fine to refer to these often-cited works but ask yourself whether you want to put them into the center of your discussion.

8c) p. 15809, l. 21 “It was found that the seasonal variation of the nighttime concentration was not due to a seasonal change in the vertical mixing, as observed for the daytime concentrations.” This is a critical issue in the discussion of the paper, and it is mandatory that the authors expand on this issue. In summer the entire boundary layer over the European continent reaches, as a whole, higher levels. Towards the night, the mixed layer height falls down, as can be seen in models (Fig. 6) and, more directly, radiosoundings. This downfall is caused by the cut-off of radiation-driven convection plus some radiative cooling closer to the ground. What remains above the ground inversion at night-time is the so-called “residual layer”, which is not equivalent to the free troposphere but composed of the remnants of the boundary layer of the previous day (e.g., Stull, 1988, An Introduction to Boundary Layer Meteorology, Kluwer Acad). This residual layer is often neutrally stable, and can contain the trace gases as well as aerosol from that preceding boundary layer. It is straightforward to see that the mixed layer height at night is not sufficient to decide whether the air at Puy de Dôme is within

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the free troposphere of not.

Therefore I recommend the authors to use other established parameters, such as moisture content, ozone, carbon monoxide, radon etc. to develop a criterion after which their night-time aerosol data can be split into different categories like “boundary-layer influenced”, or “free tropospheric”. If such results can be produced, they would be the greatest asset of the work presented.

8d) p. 15809, l. 23 “In order to provide information on aerosol physical properties that can be extrapolated to the free troposphere, we reduced our analysis to nighttime measurements.” In my opinion, the day-time measurements are equally interesting since they are a) more representative for the total radiative forcing, and b) more comparable to other ground measurements at lower levels. Night-time and day-time results could be presented side-by-side in the paper.

Language

Avoid double indexing in D_{pN} (p. 15799). Typos (incomplete list): p. 15793, Earth radiative; p. 15795, scarse.

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