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Seasonal variation of aerosol size distributions in the free troposphere and residual layer at the puy de Dôme station, France

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Abstract. Particle number concentration and size distribution are important variables needed to constrain the role of atmospheric particles in the Earth radiation budget, both directly and indirectly through CCN activation. They are also linked to regulated variables such as particle mass (PM) and therefore of interest to air quality studies. However, data on their long-term variability are scarce, in particular at high altitudes. In this paper, we investigate the diurnal and seasonal variability of the aerosol total number concentration and size distribution at the puy de Dôme research station (France, 1465 m a.s.l.). We report a variability of aerosol particle total number concentration measured over a five-year (2003–2007) period for particles larger than 10 nm and aerosol size distributions between 10 and 500 nm over a two-year period (January 2006 to December 2007). Concentrations show a strong seasonality with maxima during summer and minima during winter. A diurnal variation is also observed with maxima between 12:00 and 18:00 UTC. At night (00:00–06:00 UTC), the median hourly total concentration varies from 600 to 800 cm^{-3} during winter and from 1700 to 2200 cm^{-3} during summer. During the day (08:00–18:00 UTC), the concentration is in the range of 700 to 1400 cm^{-3} during winter and of 2500 to 3500 cm^{-3} during summer. An averaged size distribution of particles (10–500 nm) was calculated for each season. The total aerosol number concentrations are dominated by the Aitken mode integral concentrations, which drive most of the winter to summer total concentrations increase. The night to day increase in dominated by the nucleation mode integral number concentration. Because the site is located in the free troposphere only a fraction of the time, in particular at night and during the winter season, we have subsequently analyzed the variability for nighttime and free tropospheric (FT)/residual layer (RL) conditions only. We show that a seasonal variability is still observed for these FT/RL conditions. The FT/RL seasonal variation is due to both seasonal changes in the air mass origin from winter to summer and enhanced concentrations of particles in the residual layer/free troposphere in summer. The later observation can be explained by higher emissions intensity in the boundary layer, stronger exchanges between the boundary layer and the free troposphere as well as enhanced photochemical processes. Finally, aerosols mean size distributions are calculated for a given air mass type (marine/continental/regional) according to the season for the specific conditions of the residual layer/free troposphere. The seasonal variability

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in aerosol sources seems to be predominant over the continent compared to the seasonal variation of marine aerosol sources. These results are of regional relevance and can be used to constrain chemical-transport models over Western Europe.

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