Response to review comments and short comment on ACPD-12-11979-2012

The original comments are provided in black, our response is given below each comment in red.

Response to short comment from Dr. K.E.J. Lehtinen kari.lehtinen@uef.fi

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The conclusions of 'non-local nucleation' arising from analyzing figure 7, from the 'low ratio of 3nm to 6nm particle number concentrations' (Discussion, p. 11996, line 5) and 'the presence of a closed contour line around the highest number concentrations' (caption of fig. 7) may be wrong. As the contour plot in figure 7 is expressed as dN/d(logDp) (which is equal to Dp*dN/dDp), the phenomenon seen in the figure (the reddest colour is not at 3 nm but at somewhat larger sizes) is actually expected for an aerosol growing by condensation, see e.g. Lehtinen and Kulmala (ACP 3, 251-257, 2003).

Answer:

We accept that we should note that condensation can result in a closed contour and have added the following to the end of section 3.1:

"However, it should be acknowledged that a closed contour in this type of depiction of the PSD can also arise due to vapor condensation on an in situ particle population (Lehtinen and Kulmala, 2003). "

Nevertheless we wish to take this opportunity to provide a little more discussion of this matter:

A PSD expressed in terms of dN/d(logDp) is indeed expected to show higher number concentrations for larger particles. However this behavior may be not attributed just to particle growth by condensation as also stated in Lehtinen and Kulmala (2003): "There are, however some particles present, which most probably come from mixing of air".

In order to investigate this issue and quantify the contribution of condensation to the initial growth we followed the work presented in (Lehtinen and Kulmala, 2003) and extrapolated the growth rate of peak concentrations, assuming it is constant also for particles smaller than 6 nm (see Figure below). Thus the time required for particles to grow from 1 nm to 16.8 nm (i.e. the diameter of peak concentrations measured 2 hours after nucleation start) can be computed as:

$$time = \frac{\Delta D_p}{GR} = \frac{15.8nm}{4.33nmh^{-1}} = 3.65h$$

Based on this calculation we would expect the 1 nm particle peak to occur around 7:20 am (LST) which is 2 hours after the sunrise (i.e. 5:30 am LST) and thus a time of the day favorable to the photochemical production of nucleation precursors. The UHMA model, which was initialized with an unimodal particle size distribution centered on the smallest treated particles and concentration of condensable species measured during 17 May, the time required for the number geometric mean diameter to grow from 1 nm to 10 nm by condensation is always much shorter than the delay in observing substantial 10 nm particles (thus implying another mechanism – in this case the delay in erosion and entrainment was responsible).

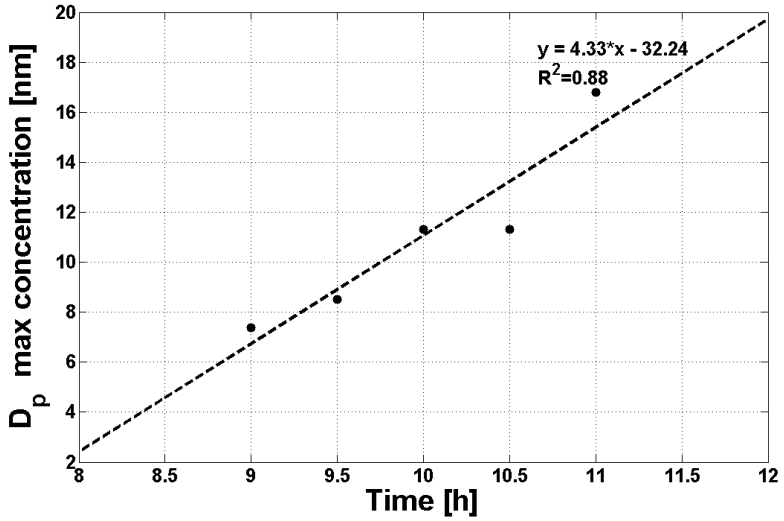

Figure 1. Temporal evolution of the diameter associated with peak 30 minute average particle number concentrations measured on May 17. The slope of the regression line represents the growth rate of particle concentration peak.

Another argument supporting our interpretation comes from the analysis of particle surface area. The sunrise at MMSF on 17 May 2008 was at 5:30 AM (LST) thus the boundary layer depth started growing after that time. If the residual layer air was free of freshly nucleated particles we would see a decrease in the surface area of 6-30 nm particles before nucleation start at the surface which is not supported by measurements (see figure below). A similar conclusion can be drawn considering the total surface area of particles in the size range 6-100 nm which present quite constant values before nucleation start, thus supporting our speculation of a major role played by mixing and entrainment from the residual layer.

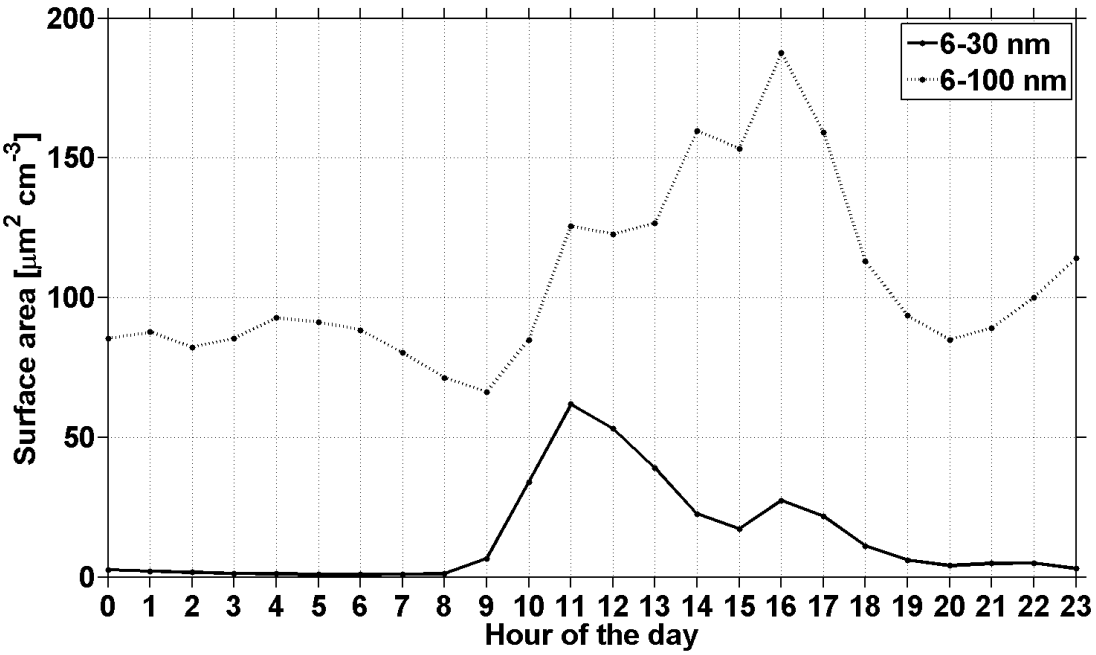

Figure 2. 6-30 and 6-100 nm particle surface area ($\mu m^2 cm^{-3}$) measured on 17 May.

We also would like to point out that Laaksonen et al. (ACP 8, 2657-2665, 2008) made aircraft measurements during a nucleation event in Hyytiälä and showed that the event started at the surface and evolved with the evolving boundary layer.

Answer:

This is a fair point – i.e. that NOT all observations imply an elevated source. We have noted this in the manuscript text (in section 1) by adding the following text:

'It should be noted that not all investigators have found evidence for an elevated nucleation source. Data collected using an instrumented aircraft and micro-light flown over the Hyytiälä site during 28 March 2003 provide evidence for a surface-based source, with subsequent vertical dispersion as the mixed-layer grew (Laaksonen et al., 2008).'