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Interactive Comment

Interactive comment on "The role of VOC oxidation products in continental new particleformation" by A. Laaksonen et al.

N. Donahue (Referee)

nmd@andrew.cmu.edu

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This is a very interesting paper that deserves to be published. The major conclusion is that, in southern Finland, new particle growth appears to be controlled by organic oxidation products associated with the forest canopy (ie, terpenes). The evidence for this conclusion is convincing. The data on the growth factors combined with the flight data demonstrating that particle growth is more or less confined to the boundary layer in Finland combine to make a very strong case.

I am less convinced by the arguments regarding the identity of those organics – that aldehydes and specifically pinonaldehyde are the major contributors to this growth mode. I have a very, very hard time believing that pinonaldehyde has a low enough vapor pressure to even be in the condensed phase under ideal circumstances at the

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low total organic aerosol loadings typical of these nucleation events, much less to overcome the Kelvin barrier challenges to growth at 2 nm. I am concerned that gas-phase adsorption artifacts may be influencing both the GC and NMR data reported here. However, this does not challenge the overall interpretation of the study, just some of the details.

P 2722 line 14: The statement in the introduction "It is suspected that the vapors causing the condensational growth of the clusters to observable sizes are low vapor pressure organic species ..." is too broad. Clearly low vapor pressure compounds are needed to sustain condensational growth of the TSCs. Organics can play that role, but sulfate certainly can as well – this has been convincingly demonstrated by Stanier et al and Qi Zhang et al (EST 2004) for Pittsburgh, as the authors discuss later in the article. Neither Pittsburgh nor Hyytiälä is globally representative – discovering where sulfate and where organics play this vital role in new-particle growth is a major need.

P 7825 line 2: Please state the vaporiser temperature as well (600 °C?). This influences the organic mass spectra by adding a thermal pre-heat to the electron impact ionization.

p 7828 line 15: I have a very hard time believing that pinonaldehyde has much to do with the growth of sub-10 nm particles at low total aerosol mass. We see pinonaldehyde in the gas phase, where it belongs, using PTRMS (Presto EST 2006 – Supplemental Material). The saturation concentration of pinonaldehyde is 1000 μ g m $^{-3}$ or more. However, it can adsorb to quartz filters, leading to a positive artifact in filter sampling. This artifact would, however, be correlated with the first-generation a-pinene SOA products in the air and thus might serve as a useful surrogate for the much lower vapor pressure material that must be needed for new particle growth.

The only reasonable counter argument to this point I can see would be for the pinonaldehyde to react heterogeneously on the particles to form much less volatile oligomers. However, there are two problems with this. First, once the pinonaldehyde

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reacts it is hard to understand how it would then be detected as the monomer (reversible oligomerization just won't do it, in my opinion). The other problem shows up in Figure 4 of our supplemental material again – at least in the chamber, the pinonaldehyde appears to be perfectly stable – if it were engaged in a heterogeneous reaction associated with particle growth we would expect loss.

p 7828 line 22: 23.5%?? That is awfully precise. I think one significant digit, and not three, is in order here.

p 7830 line 5: If the size range of the total particle measurements was indicated, I missed it and can't find it easily again. Please state it here.

Table 1: There is an extra word ('and') in the caption. It would be useful to know the total organic aerosol mass (OA) in these samples too in order to get an idea of the expected partitioning.

Fig 2: I find the linear fit here misleading. It is clearly correct to assert that the periods of highest MTOP are all associated with high EtOH growth factors, but there are plenty of high-growth points at low MTOP too – it appears that high MTOP leads to high EtOH growth, but that other things can too.

Fig 4: Please resort the legend so that high Theta is at the top – this confused me for a second! Also, virtual potential temperature (including water vapor buoyancy) is typically a better measure of boundary layer stability. I doubt that influences the qualitative point here, though.

Fig 5: It would be interesting to reproduce the turbulent mixing height line here – the correlation (aside from the general sign of the trend) is not obvious to me.

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