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Comment

***Interactive comment on* “The link between atmospheric radicals and newly formed particles at a spruce forest site in Germany” by B. Bonn et al.**

Anonymous Referee #1

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New particle formation in the atmospheric boundary layer is a scientifically challenging research field. Due to potential feedback mechanisms especially new particle formation involving biogenic precursor gases are of special interest. Recently a lot of interest has been given to the involvement of organic species in the nucleation process, either directly in the early stages of cluster formation or at a later stage when the nanometer sized particles are growing. Exactly this is the topic of the manuscript from Bonn et al. Therefore the subject of the manuscript is completely suited to be published in Atmospheric Chemistry and Physics. However, the way the authors present their main research statement— as a proof of a direct involvement of large peroxy radicals

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and stabilized Criegee radicals in new particle formation at the measurement site – is scientifically not convincing and stays on the level of a working hypothesis. The general idea that organic compounds, which derive from the oxidation of VOCs (including biogenics), are involved in new particle formation is certainly a valuable conclusion from the data presented in the paper, however, the proof that particle formation follows exactly the presented nucleation scheme (as already mentioned above in a complex manner with large organic peroxy radicals and stabilized Criegee radicals playing the central role) is simply missing. There is no question that these radicals play an important role in the oxidation chain of VOCs and that they are essential intermediates to understand the formation of oxidized and therefore lower volatile products, however, the paper misses to present clear evidences that atmospheric radicals are directly involved in particle formation – and not the stable end products of the peroxy radical chemistry in the gas phase. Again, in my opinion the data presented are very valuable and worth to publish. I suggest that the authors discuss the results in terms of evidence or proof of the involvement or biogenic VOC oxidation products (whatever these products are: radicals, larger peroxides, permutation products of radical chemistry, highly oxidized or extremely low volatile species produced from radical intermediates. . . .) in new particle formation without announcing that we know now how exactly the mechanism is.

Therefore, I suggest major revisions to the manuscript before it is published in ACP.

Some minor comments:

Page 27504: Why are small particles (1.5 nm) ‘chemically’ unstable ?

Page: 27506: What do you mean with ‘. . .split up of larger ones . . .’. Where and when is that happening ?

Page: 27507: The way the authors discuss is often scientifically not precise: Surface production and the thermodynamic consequences of the formation of a new phase result in the Kelvin effect, however, they are not ‘the Kelvin effect’.

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Page 27508: 'vegetated boundary layer' ?

Page 27509: The whole sequence discussing the different involvement of smaller and larger RO₂ radicals is very weak. I understand that the authors would like to avoid contradictions to previous observations about the effect of isoprene, however, this is done with a combination of hypotheses and statements about peroxy radicals and cluster chemistry ('seal the reactive sites' ??) which is not scientifically sound !

Page 27514: The reaction rate constant of sCI and NIM: Where does it comes from ?

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 27501, 2013.

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