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Interactive Comment

Interactive comment on "Ambient new particle formation parameter indicates potential rise in future events" by B. Bonn et al.

Anonymous Referee #1

Received and published: 5 March 2009

The manuscript describes a nucleation parametrization based on reaction of biogenic VOC (sesquiterpenes) with ozone, stabilized Criegee radicals being a key intermediate. As new aspects a temperature dependence of "terpene" emissions and a proxi for OH radicals (UV B) was included. The resulting nucleation parameters and their organic cores are compared with observations number concentrations of 10-12 nm particles at the Taunus (Germany) and with number concentrations of 3-6 nm particles and event frequency in Hyytiälä.

The increasing role of organics in the nucleation of particles, possibly of the cost of the importance of sulfuric acid is currently heavily investigated and discussed. From this point of view the manuscript seemed to be important and timely. However, the manuscript is highly speculative in the development of the nucleation parameters and



the comparison to the field observation does not nail down the uncertainties and speculations. From this point of view the future predictions within the paper thus seems extremely wage.

I have mayor concerns:

Also not always explicitly mentioned - sometimes the authors use the notation "terpenes" which could include monoterpenes MT - the core of the mechanism are stabilized Criegee radicals of sesquiterpenes SQ and the anticorrelation of the nucleation threshold of SQ (caryophyllene) with absolute water vapor, as observed by one of the authors in a laboratory study.

The summer minimum in nucleation in Hyytiälä where water concentration ares highest, as well as the fact that humidity often is lower on event days, is used as an argument in favor of the SQ - mechanism. Is the connection between the microphysical process of scavenging Criegee radicals and a multidimensional "metorological" parameter like atmospheric humidity sufficiently supportive ? Lyubovtseva BER, 2005, discuss that more critical. Moreover, are the leverages in the water dependence of SQ nucleation threshold and summer water concentration in July (nucleation minimum) and August (nucleation peak) sufficiently strong to explain non-nucleation / nucleation ?

If ozonlysis of SQ is the core of nucleation why one needs OH (UV B) to be included as a proxy?

If sesquiterpenes are key, is the pool emissions formulation appropriate ? Even if SQ may have a similar T dependence as MT they have more variable standard emission E0 and possibly a light dependence. Moreover in their paper in 2005, Tarvainen et al concluded that the annual cycle of SQ does not match the spring nucleation, but possibly the late summer maximum of nucleation. That seems to be in contrast to your nucleation parameter, which fits in spring and fails the August maximum.

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Are the Taunus measurements suited to separate nucleation and condensational growth in the the sense of veryfying a microphysical mechanism? You already missing one of your self- defined key parameter, the UVB measurement. I also don't see what is meant by agreement between NP'S and the particle number concentration. Particle number vary much more than the NP's. The relation of the time shift of NPA and NPA(org) and its effect on two nucleation events is not so clear to me. I can only see this effect on the last day and maybe on day 133, although the two parameters are always shifted with respect to each other.

The Hyytiälä data have two maxima but you can only predict one, the explanation and the information in the appendix do not solve that problem. Aldehydes and the NP according to Bonn 2008 peak later in the year than August

Based on this concerns, I find this paper too speculative and it is not suited to be published in ACP in current form . The paper needs better support by observations (and argumentation) along above concerns before publication in ACP. In any case future prediction should be avoided at the current stage.

Moreover the manuscript is difficult to read at some instances The figures 1 and 2 are of insufficient quality. And the figure caption in figure 3 seems to be wrong.

Detailed points:

page 675, line 27: the sentence does not make sense to me.

page 676, eq 3: stab. fraction is not explained

page 677, eq 7: what is the physical dimensionality of these NP's ?

page 678, line 2: this year ? 2008 !

page 679, line 14 ff: this is difficult to follow.

page 679, line 22 f: How can the difference be caused by the MT emissions, these do not enter explicitly the NP's ? Or is it the different T which drives the pool emission

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parametrization?

page 680, line 11: you are not applying the "Guenther" algorithm, but only the "pools part". Guenthers light intensity might even help for SQ, if it would be known !

page 683, line 24: What are severe predictions ?

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