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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 #2

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The manuscript of Bonn et al. reports on a new nucleation parameter that represents the role of biogenic compounds in forming new particles. This new parameter is used in modeling particle number (< 12 nm) and frequency of nucleation events. The modeling results are compared with data from two boreal forest sites, one in Germany and another in Finland. The new parameter is a function of ozone and water vapor concentrations, biogenic emissions, and temperature. Given that those inputs are projected to change with changing climate, Bonn et al. use the new parameter to predict nucleation under future conditions using projected changes in the listed inputs. Based on their predictions, Bonn et al. conclude that nucleation frequency will increase under projected future conditions over boreal forests. There is still much to be understood about the role of biogenic compounds in new particle formation and advances to be made in modeling





nucleation events, and thus the manuscript is very relevant. However, the manuscript lacks clear description and critical discussion of the new parameter and modeling results obtained. Application to projected future conditions, in which further uncertainties are introduced, appears unsupported at this time. It is suggested that the manuscript be significantly revised before publication in ACP. Specific comments are provided below. Some of the comments may be the result of misunderstanding/misinterpretation. It is suggested that during revision, the authors seek assistance in editing, possibly from a native English speaker.

Specific comments

Section 2: The origination of the new parameter organic NPB is not sufficiently described. The authors state that in addition to the terms in the parameter organic NPA, "terpenes are needed". From this statement, it is unclear why terpenes were not included in organic NPA. Additionally, there is no description of where the term representing terpene emissions comes from, including a definition of beta. Including units may help clarify the new term. The use of units also is recommended for the two equations with the UV B term (Eq. 7).

p. 677-678: The authors note that when data are not available, UV B = 1% incoming solar radiation and discuss the implications of that assumption in the results. It is suggested that the UV-B data that are available be included (at least the range of values), so that the effects of this assumption can be better quantified. Other assumptions are also discussed including the temperature dependence of emissions and terpene selection. Regarding the former, the authors discuss using two different assumptions regarding temperature; however the results of those assumptions are only briefly discussed, and in most of the discussion, it is unclear which results are being shown. Regarding the latter, it may be useful for the authors to include some discussion of the variability in reactivity of different terpenes, including some range of rate constants and stabilized fractions, and the potential effects that variability would have on their results.

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p. 677, line 18: The authors state that in some cases terpenes are assumed to present in "sufficient amounts". It is not clear what constitutes sufficient amounts.

Section 3.1: Overall, this section is hard to follow. In lines 6-8, the authors state that the Taunus observatory is "dominated by secondary sources", which "is most effective at winds from remote regions", "in contrast to Frankfurt". It is not completely clear what the authors mean here. It is assumed that they mean that the emissions are predominately biogenic rather than anthropogenic, but there is an implication that wind direction is important though not shown/discussed further in results.

In lines 13-17, the authors discuss a shift in the parameter organic NPA from "the nucleation parameter". It is assumed that the authors mean from the nucleation parameter NPA. The authors attribute the shift to the daily solar cycle and state "If this is apparent..." two maxima "are visible". It is suggested that the word "apparent" be replaced and that the use of "if" and "are" be reconciled (i.e., if/would be or is/are). In line 16, "to" should be "two".

More generally, it is not clear why the authors do not plot or discuss NPB, since it is assumed that NPB is the new parameter being introduced in the paper. Further, while the authors conclude that generally good agreement is achieved, there are several features (nucleation events) that are not predicted, and the reasons for the varying degrees of agreement (e.g., why the results using one parameter or the organic part of one parameter more closely matches the data) are not discussed here or for subsequent sections.

In lines 19-22, the authors conclude that NPB more closely matches data, and thus there is a notable contribution from biogenics. While NPA does not explicitly include terpene concentrations, doesn't NPA also represent a contribution from biogenics?

In line 22, the authors address the difference in magnitude of the nucleation parameter. It is not clear whether this is parameter NPA or NPB. They attribute the difference to difference in terpene emissions. Is it the magnitude of emissions or the types of 9, S420–S424, 2009

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emissions? If the former, it may be helpful to include the values of the emissions so that their impact on the nucleation parameter can be better understood.

In lines 23-26, the authors discuss the organic part of NPA indicating a clear seasonal change, however organic NPA isn't shown for Hyytiala. It is not clear if the authors are referring only to Taunus or to both sites. If Taunus only, it is suggested that the discussion be moved to the preceding paragraph; if both, it is suggested that organic NPA for Hyytiala be shown.

The last sentence of section 3.1 discusses the suppressing effect of water vapor on nucleation. It would be helpful if the authors reported humidity data to further support the discussion and conclusions here and throughout the manuscript.

Section 3.2, lines 7-19: The authors state that the mismatch between predicted and observed nucleation frequency in August, using the parameter organic NPA, could be due to "neglect of the organic hydrocarbon contribution". As mentioned above, doesn't organic NPA represent the organic contribution? Are the authors referring to the need for an emissions term? This doesn't necessarily seem to be the case, since organic NPB also misses the August peak in nucleation frequency. The explanation in lines 10-14 suggests that the authors are referring to the mismatch with NPB, but that mismatch has not been discussed. Aside from the confusion as to which of the parameters the authors are discussing, the explanation for the temperature hypothesis is difficult to follow. For example, the authors state the care should be taken using the algorithm of Guenther et al., 1995, which was obtained at warmer temperatures. It seems warmer temperatures would lead to higher emissions, which would lead to increased nucleation frequency. It may be useful here to include results obtained using a different assumption regarding temperature and emissions, such as described in Section 2.

In lines 9-10, the authors also state that the mismatch could be due to the suppressing effect of trace gasses on nucleation. Based on the discussion beginning on line 20, it seems the authors are attributing the nucleation peak in August to formaldehyde, and

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the suppression of that peak in June and July to the loss of the SCI intermediate to organic acids. The mismatch in August would then seem to be related to the inability of the nucleation parameter to represent the contribution of formaldehyde, and the closer match in June and July the ability of the parameter to represent the suppression of the SCI intermediate. However, as noted by the authors, that reduction is not included in the parameter. This hypothesis could use further discussion/clarification.

Section 3: The authors suggest that the new nucleation parameter, organic NPB, represents the annual pattern in nucleation events. It is suggested that Figure 3 be revised to more clearly show the suggested pattern. Additionally, while it can be inferred, the authors do not define "scaled" or "Spring" NPB, and do not discuss Spring organic NPB in this section. Because there is a top and bottom figure for each numbered figure, it is sometimes difficult to follow which figure is being discussed. Perhaps Spring NPB was discussed previously. It is suggested that the authors always refer to the figure number and top or bottom.

Figures: It is suggested that the authors use larger text and thicker/darker lines in Figure 2. Figure 4 needs a caption; currently it is the same as for Figure 3.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 673, 2009.

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