Interactive comment on “On the roles of sulphuric acid and low-volatility organic vapours in the initial steps of atmospheric new particle formation” by P. Paasonen et al.

Anonymous Referee #1

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This manuscript aimed to investigate the influences of sulphuric acid and organic vapors on atmospheric new particle formation rate. Though this is an important and fascinating topic, the reviewer feels that this work adds limited contribution to the field. The correlation between new particle formation rate and sulphuric acid has been systematically examined before (e.g., Weber et al., 1997, Riipinen et al., 2007; Kuang et al., 2008). Elucidating the role of organic compounds in atmospheric nucleation would be a significant development; however, the scientific approach and applied method in this manuscript need further justification. Rather than directly measuring concentrations of low-volatility organic vapors, empirical methods were used to estimate them. Among many assumptions made while estimating organic compound concentration,
using equation (1) implies that the uptake of organics by new formed particles is purely a physical mass transfer process, i.e., condensation. Though it has been observed that the uptake of H2SO4 by new formed particles can be modeled as the condensation process (e.g., Sipila et al., 2010), the uptake of organics are most likely limited by surface reactions rather than by mass transfer. Accordingly, equation (1) would underestimate the concentration of organic compounds. The estimated organics concentrations for four sites are $\sim 10^7$ molecules/cm$^3$ (Table 1), which are not surprisingly several orders below the concentrations directly measured during a recent campaign. Therefore, the proposed correlations between new particle formation rate and organic compounds, which are based on those estimated organics concentrations, are highly questionable.

The proposed homomolecular organic vapor nucleation, $K[\text{org}]^2$, is primarily motivated by the data from Hohenpeissenberg site. It implies that organics alone (without H2SO4) can trigger atmospheric new particle formation events. The time dependent H2SO4 concentration along with particle size distributions should be included to justify this argument. In addition, only two nucleation events from this site were studied in this manuscript. The authors mentioned that “measurements at HPB are continuously running and corresponding data should become available soon”. More nucleation events will certainly help to further evaluate the proposed mechanism.

The authors assumed that the same organics responsible for particle growth are also responsible for atmospheric nucleation. This needs further justifications.

In summary, additional work are required before the manuscript can be considered for publication at Atmospheric Chemistry and Physics.

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