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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Inversely modeling homogeneous H_2SO_4-H_2O nucleation rate in exhaust-related conditions" *by* Miska Olin et al.

Anonymous Referee #1

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The manuscript entitled "Inverse modeling homogeneous H2SO4-H2O nucleation rate in exhaust-related conditions" by Miska Olin et al. investigates particle formation in typical vehicle exhaust conditions both experimentally and theoretically. Combined measurements of particle number concentration and size distribution by PSM and nanoSMPS are taken to determine nucleation rate as a function of sulfuric acid concentration (measured by CI-API-ToF-MS and ion chromatography), RH and sulfuric acid saturator temperature. Modeling of the exhaust sampling system together with the aerosol model CFD-TUTMAM lets the authors conclude that binary sulfuric acid-water nucleation is unlikely the dominant mechanism in the particle formation of real-world driving situations. Instead, additional compounds such as hydrocarbons are claimed necessary to explain the observed particle concentrations. While I was positive about

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Discussion paper

this manuscript initially as this topic is of high relevance to the ACP readership and inverse modeling is also becoming more and more popular nowadays I regret to say that I cannot recommend publication in ACP at this stage.

Beginning with section 2 reporting on the experiments I got the feeling that the authors themselves have no faith in their own experiments/results. Two different instruments for the measurement of sulfuric acid concentration yield substantially different results and it is not clear what the reason for this is. Similarly, the combined size distribution measurements from PSM and nanoSMPS do not produce satisfying agreement in the overlapping size regions. My guess is that the used sizing instruments do not provide the features needed to capture correctly the time scale of the particle dynamics taking place in this exhaust type experiment. To me it seems critical that the experimental data need a much higher confidence level to allow comparison to the modeling results. The concluding section clearly documents the reasons for my concerns about this study. It nicely summarizes what has been done and what problems were encountered but it does not come up with major and firm scientific advances that I would expect from a paper in ACP. The main conclusion seems to be the inappropriate binary H2SO4-H2O nucleation mechanism and that other vapors would be needed for multicomponent nucleation. But this claim is firstly vague, and secondly, not unexpected and thus limited in novelty. In the end I feel that this manuscript in its present form may better fit a technology focused journal but it is certainly not suitable for ACP.

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