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

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We thank the referee for reading the manuscript and for bringing up the important issues of the manuscript.

We agree that the measurement of sulfuric acid had high uncertainty and the two measurement approaches (CI-APi-TOF and IC) yielded different concentrations (varied within 2 orders of magnitude) due to high diffusional losses when sampling with the CI-APi-TOF. In general, measuring sulfuric acid concentrations is still always a challenging task to date; the major contribution of the uncertainty arising from high losses of sulfuric acid onto the sampling lines before the chemical ionization inside the CI-APi-TOF. For these reasons, we did not use the measured sulfuric acid concentrations

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in the simulations; instead, the sulfuric acid concentrations were obtained through inverse modeling by using the measured particle size information. Inverse modeling of the vapor concentrations is possible due to the condensational growth of particles, and the estimation of condensing vapor from the observed growth is widely used method in atmospheric nucleation studies. These simulations could have been performed even without the sulfuric acid measurement still producing the same output. We provided the results from the sulfuric acid measurement as additional, supporting information on the sulfuric acid concentrations for the manuscript. Based on the referee's response, we see that the presentation of the measurement of sulfuric acid would have been better suited as a supplement for the manuscript.

The case of the disagreement of the overlapping size regions measured by the PSM (and the CPC) and the Nano-SMPS is observed elsewhere also (Kulmala et al., 2013; Alanen et al., 2015; Rönkkö et al., 2017). It is caused by drastically underestimated particle concentrations at the smallest particle sizes measured by the Nano-SMPS due to high diffusional losses of small particles inside the device, due to low charging efficiency of small particles, and due to the inversion algorithm which favors features from a log-normal size distribution. It should not be related to the time scale of the particle dynamics in an exhaust type experiment, because this disagreement has been seen in atmospheric measurements also (Kulmala et al., 2013).

The referee found the main conclusion of the manuscript being that binary H2SO4-H2O nucleation is unlikely to be the mechanism in vehicle exhaust, with the opinion that this is not a novel conclusion. We would like to state that the main output of the study was meant to present the binary H2SO4-H2O nucleation rate as a function of sulfuric acid concentration, water vapor concentration (or RH), and temperature in high sulfuric acid concentrations and temperatures, based on experimental data. Such a rate is currently not available for the relevant temperature and concentration ranges, and therefore we consider that our result is novel and of importance to atmospheric science and thus the manuscript should be suitable for ACP. We agree that the inappropriateness of binary

H2SO4-H2O nucleation mechanism occurring both in the atmosphere and in vehicle exhaust is not a novel finding (Saito et al., 2002; Vaaraslahti et al., 2004; Meyer and Ristovski, 2007; Kerminen et al., 2010; Sipilä et al., 2010); thus, it was not meant to be the main conclusion, but act as additional information and stronger support to the conclusion of former studies related to the nucleation mechanism occurring in vehicle exhaust. It is an additional result of our study; our experiments are also, to our knowledge, the first ones that show directly and experimentally that the formation rate of particles at high sulfuric acid concentrations and in the absence of additional compounds is lower than in H2SO4-containing vehicle exhaust. In light of the referee's comments, we agree that this aspect of our study was highlighted too much in the manuscript to give the feeling of it being the main conclusion. Should we have the opportunity to prepare a revision, we will take the strong criticism of the referee on board.

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