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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 #2

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New particle formation in vehicle or other exhaust flows as well in atmosphere, is one of the least understood aerosol processes. Regarding vehicle exhaust, the measurements indicate that although sulfuric acid participates in the production of volatile particles, it cannot explain the measured particle number size distributions. So, volatile and/or semi-volatile condensable vapours other than sulfuric acid are required to explain the measurements. This is the general story of the exhaust nanoparticles science. And the questions are: Which are these other vapours? And, which are the alternative mechanisms? Several studies, both experimental and theoretical, have been conducted to answer these questions and, although the levels of instrumentation and of modelling sophistication and complexity have significantly increased over the recent years, this is not the case for the level of a clear understanding of the processes. At

least this is my perspective, and I think that this is also the view of the authors, at least as indicated by their conclusions (page 31/38, lines 15-17): "[. . .] This indicates that H2SO4 and H2O cannot fully control the nucleation process; instead, other compounds, such as hydrocarbons, existing in real exhaust are likely".

Irrespective of the above general statement, the article is interesting, it can contribute to further research and can be considered for publication in the journal. Nevertheless, I think that the authors should be more convincing when presenting the need for the proposed study. Below, my detailed comments:

Title – good, reflects the content of the paper

Abstract – good, describes the essential information in the work.

Introduction – It is rather well written and in sufficient detail. Very good presentation of the work already done in the field. I have some remarks, however: âĂć Pages (2-3)/38: It will also be useful if the authors reconstruct slightly the text. More specifically: it would be better for the reader if the authors had continued the description of binary homogeneous nucleation (BHN) after line 33 in page 2 with the text of page 3 (lines 6-12: "The derivation of the CNT contains, however .... with experimental nucleation rates") and had started a new distinct paragraph with the details of the other nucleation mechanisms (i.e., from "Conversely, the nucleation rates of the other nucleation mechanisms" up to "thus, a constant coefficient cannot be used." An then from (page 3 line 12) : "The nucleation exponents, n, for H2SO4 obtained" up to "much lower than the theoretical exponents (n > 5) (here, references are needed)." âĂć Page 3/38, line 1: Explaining equation 1, the authors write "[...] and n is the nucleation exponent presenting the sensitivity of [H2SO4] on J." It would be useful for the reader to understand better sensitivity if the authors add that, in theory, the nucleation exponent n represents the number of nucleating molecules in the critical nuclei. âĂć Page 3/38, line 15: "The first attempts to obtain the nucleation. . .." This is not exact. Vouitsis et al. (Modelling of Diesel Exhaust Aerosol during Laboratory Sampling. Atmos. Environ.39, 2005) showed that the

barrier-free nucleation scheme, where clusters are always stable against evaporation, could predict the nucleation mode particles concentration rather well for low sulphur fuel (<10 ppm), whereas a nucleation rate proportional to the square of sulphuric acid saturation vapour pressure was more appropriate for high sulphur fuel (250 ppm).

The research question is stated in a somewhat unclear and contradictory manner. The authors note (page 4/38 lines 9-14) that their research focuses on “pure H2SO4-H2O nucleation instead of nucleation associated with some unknown compounds existing in real vehicle exhaust“ on the basis of the very low level knowledge of the H2SO4-H2O nucleation mechanism; however, some lines further (lines29-29) they state that “The formulation obtained from this study helps in finding the nucleation mechanisms occurring in real vehicle exhaust or in the atmosphere, etc”. I think that the argument must be rephrased in a more proper statement.

Materials, methods, discussion - The methodology presented in the manuscript and the analysis provided are both accurate and appropriate. The experimental section is robust and the methods applied are presented in sufficient detail. The observations made are discussed comprehensively and the data presented are adequate to support the conclusions. The same is valid for the simulation section of the study. The authors have developed a very interesting model which accounts for the flow and the temperature field of the sampling system, for the solution the aerosol processes, for the correction of water amount present in the particles and for diffusional losses in the sampling as well as for the detection efficiencies of particle counting devices. One comment only: in two cases, (page 30/38, line 8 and page 3/38m line 18), the authors write “The obtained exponent nsa = 1:9 is in agreement with the former studies”. However, they do not refer to any study specifically, unless if they mean the article of McMurry and Friedlander (1979), on which they referred in the first case. If this is true, they must be more specific in relation with the observations of McMurry and Friedlander and their connection with the results of this study.

Conclusions – I have a similar and related with the above comment in the introduction

section concerning the scope and clarity of the study. The authors write: “[. . .] nucleation rate obtained in this study helps in finding the currently unknown nucleation mechanism occurring in real vehicle or power plant boiler exhaust or in the atmosphere”. The authors must clarify better how this help is provided.

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