Atmos. Chem. Phys. Discuss., 15, C1113–C1116, 2015
www.atmos-chem-phys-discuss.net/15/C1113/2015/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Model studies of volatile diesel exhaust particle formation: organic vapours involved in nucleation and growth?" *by* L. Pirjola et al.

Anonymous Referee #1

Received and published: 25 March 2015

Pirjola et al. report their work aiming to model the behaviour of the aerosol particle number size distribution in engine exhaust emissions within the measurement set-up. The authors test several nucleation mechanisms and compare how well the output from the measurement set-up can be reproduced with the model when the different nucleation mechanisms and assumptions on condensing vapours are applied. Unfortunately, the manuscript suffers from unsatisfactory level of English language. Otherwise it is, however, interesting and the results are certainly valuable for the audience of ACP. If the authors manage to improve the English in the manuscript and respond to the few specific comments below, I will recommend publication in ACP.

Full Screen / Esc
Printer-friendly Version
Interactive Discussion
Discussion Paper

Specific comments:

The authors state that the measurement set-up has been shown to represent the real-world processes relatively well (page 4224). According to Fig. 3c the condensation sink in the end of the chamber, >2.5 s after the emission, is between 2 and 7 sˆ-1. The typical atmospheric conditions very rarely reach 0.1 sˆ-1. Are the values in range 2 to 7 sˆ-1 realistic to roadside values or one to two orders of magnitude higher? If it is not realistic, how much would this affect the comparison between the model, measurements and the real-world conditions?

The authors show that the HET nucleation mechanism can be adapted to all the measurement conditions with the same nucleation coefficients and COV concentrations, whereas the other mechanisms cannot. In the current version, this important finding is quite much buried under all the other results. I would suggest e.g. reforming Table 2 to a figure, where the required nucleation coefficient values and COV concentrations for the different nucleation mechanisms are presented as a function of GSA. This figure, and few more sentences on its significance would clarify the conclusions significantly.

Biofuel/diesel is mentioned only in the last sentences of the abstract and of the concluding remarks. There should be some analysis and/or sentences with references also in the text to back up the conclusions. Currently, it seems like the related part of the manuscript would have been excluded from the submitted version.

Technical comments: Many of these are related to the language, but as I am not native English speaker, not all the suggestions are necessarily correct. The authors should review the whole manuscript in terms of the English language.

- P. 4220 line 2 “during exhaust cools and dilutes” does not sound good, perhaps “while the exhaust cools and dilutes”/”during cooling and dilution of the exhaust” - P. 4220 line 8: “running with low fuels sulphur content (FSC) FUEL” - P. 4220 lines 23-24: give reference - P. 4220 line 24: "due to the tightened" would be better a bit later, after "...have been reduced" - P. 4221 line 11: pPDF should probably be pDPF - P. 4222 line 4:

ACPD
15, C1113–C1116, 2015

also sulphuric acid - organic vapour nucleation should be mentioned, e.g. Riccobono et al., 2014 - P. 4222 lines 12-17. The sentences should be recoupled: “Recently... into two groups. Some of them are process models... in this paper, whereas some are computational...” - P. 4222 lines 21-25: You have not used the word “barrierless” so far, please clarify and give reference. More importantly, in their conclusions Vouitsis et al. state that “it is shown that a nucleation rate without an energy barrier should be considered for qualitative stable predictions”, and in their Results-section that with high FSC fuel barrierless nucleation scheme overestimates the nucleation mode concentrations (and saturation ratio scheme works adequately) whereas with low FSC fuel barrierless nucleation is more appropriate. In this manuscript the authors have mixed these to the opposite. - P. 4226 lines 15-16: which CIMS measurements are referred to here? - P. 4226 lines 18-20: is it so that the adipic acid in the model condenses on all particle sizes and nucleates? What does it then mean that the thermodynamic properties are taken from the literature? - P. 4227 line 12: if adipic acid is assumed to participate in heterogenous nucleation and growth from 1.5 nm on, what is the difference between it and ELVOC? Furthermore, ELVOC is a term, which is typically used for highly oxidized C10 or C20 species. It also sounds that ELVOC would be more volatile than condensable organic vapour (COV). I suggest to use another term in stead of ELVOC, e.g. COV with some sub-index. - P. 4228 line 5: is the final exhaust temperature equal to dry air temperature, 303 K? - P. 4228 line 9-10: either “Similarly to Lemmetty et al., dilution is modelled...” or “According to Lemmetty et al., dilution can be modelled...” - P 4228 line 15: either “all” or “only” to be removed - P. 4231 line 10: “The activation coefficient and COV concentrations were varied”. Wasn’t the case that the coefficients A and K as well as COV concentration were taken as free parameters to find the best correspondence between measurements and the model? At least this is what P4231 line 21 looks like: “obtained coefficients A and K vary strongly” together with P4233 line 14-15, where the COV concentration becomes quite constant. - P4231 line 12-14: “height of the nucleation mode” would be better as “nucleation mode particle concentration” - P4231 line 27-28: some reference is needed if the difference between measurement and model

ACPD
15, C1113–C1116, 2015

is suggested to result from the underestimation by SMPS. Do the authors suggest that all SMPS-measurements underestimate the concentrations of 3-7 nm particles by roughly an order of magnitude, which is the difference between the measurement and the model in Figs. 4 and S3? - P4232 lines 4-5: “It is not expected that the activation coefficient A was not constant but varied in range. . .”? Does this mean that the authors believe it is constant in reality, but varied the value in model to see the effect in the model output? - P4234 line 7: does “other organic vapours” now refer to other than the nucleating vapour? - P4234: are both COV and ELVOC in MAFOR capable to nucleate as in HET mechanism? - P4235 lines 1-2: Is this based on previous measurements? If so, please give the reference. - P4236 some terms and sentences are unclear: lines20-22; line 24 “volatile nucleation mode”; - P4237 line 5: nucleation drops -> nucleation rate drops - P4237: here is applied a term “non-volatile nucleation mode”, earlier “core mode”, in figure captions “non-volatile core mode”. Please, harmonize. - P4239: line 8: cDPF? Previously pDPF is mentioned. - P4240, line 15: the time at the end of the simulation, 2.7 s, should be mentioned - P4240, line 18: CCN has not been mentioned earlier. - P4240, line 20: the proxy? - P4240, line 28; 2007 is not too recently any more - P. 4235: Figure and caption, EJ = ED? - P. 4257 on: the nucleation mechanism applied for the figures from here on, apparently HET, should be mentioned in the legend.

References: Riccobono et al., Science Vol. 344 no. 6185 pp. 717-721 DOI:10.1126/science.1243527

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 4219, 2015.