

Interactive comment on “Influence of fuel ethanol content on primary emissions and secondary aerosol formation potential for a modern flex-fuel gasoline vehicle” by Hilikka Timonen et al.

Anonymous Referee #3

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SUMMARY

In this work, Timonen et al. determine the influence of three different ethanol contents (E10, E85, and E100) on the primary emissions and secondary aerosol formation from a flex-fuel gasoline vehicle that passes EURO5 standards. Tests were conducted during a New European Driving Cycle (NEDC). While much of the work was presented as an average over the entire NEDC, the temporal variation in both chemical composition and variations in the size distributions were also explored. Aging of the exhaust was done in a Potential Aerosol Mass (PAM) chamber, which can simulate atmospheric aging on the timescales of days and with a high time resolution, unlike batch chambers. Aerosol characterization was achieved with a Soot-Photometer Aerosol Mass Spec-

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trimeter (SP-AMS) and an Engine Exhaust Particle Sizer; gas phase characterization was achieved with a FID, FT-IR, and GC.

In this work, the authors found that as the ethanol fuel content increased, the primary emissions of particulate matter (PM), BTEX (Benzene, Toluene, Ethylbenzene, and Xylene), the contribution of refraction black carbon (rBC) to the total PM, and the potential to form secondary PM all decreased. The concentration of total hydrocarbons as measured by FID, however, increased.

Overall, this work is sound and brings additional knowledge on the effect of fuel composition to secondary aerosol formation. There are, however, several general and specific that should be addressed increase the validity of the results and the effectiveness of the paper. Furthermore, there are many outstanding grammatical and organization errors that should also be addressed before publication.

GENERAL COMMENTS

(1) It is mentioned a couple times in this manuscript that losses through the PAM have been characterized both in general and for this system (Page 6, Line 1). The validity of this work would be greatly enhanced if these losses could be well-characterized applied to the data. This would also help interpret whether a great degree of fragmentation was causing loss of aerosol mass.

(2) How valid is it to compare the E10 and E85 PAM results to the E100 results? Looking at Figure 8, the peak aerosol production for E10 and E85 was during the CSUDC portion of the NEDC. Thus, it seems as if comparing the 1-day aging of E10/E85 to the 0.02 day aging of E100 results should either be better justified or entirely omitted from the discussion of secondary PM formation.

(3) One interesting result from this work is the chemical composition of the exhaust as function of aging. While the authors touch upon this in Figure 6, it would seem that the AMS has a much better ability to determine well-known aerosol classes through

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positive matrix factorization. Is this possible with this data set? Furthermore, is this possible as a function of time during a NEDC run, or is the time resolution not sufficient? Such work could greatly enhance the impact of this paper.

SPECIFIC COMMENTS

Page 5, Line 20: Why was deionized water added to the E100 fuel to adjust water content to 4.4% m/m?

Page 6, Line 5: Why was the PAM not placed after the secondary dilution system? It seems as if that would be the most realistic in terms of the partitioning of semi-volatile organic compounds.

Page 7, Line 13: What is the reason for this difference in collection efficiencies?

Page 7, line 15: What is the significance of 90 nm for agglomerates if the restriction set by the aerodynamic lens is 50 nm?

Page 7, line 15: Furthermore, does the soot vaporizer in the SP-AMS have a lower-detection limit like the SP2? If so, that should be explicitly stated here.

Page 10, Line 24: Why are the C_xH_yO_z fragments going down with PAM aging? Is this evidence of fragmentation?

Page 10, Line 30: Why are these losses not accounted for in this study?

Page 11, Line 28: The phrase “close to zero” has no physical meaning in this case. Please change to “below the detection limit of our fuel analysis” and indicate what that detection limit is.

Page 13, Line 33: If the total HC from GC, FT-IR, and HPLC, then why do the authors believe the yield is 30% lower than predicted?

Page 15, line 7-9: Why are small organic particles ruled out of this situation?

TECHNICAL COMMENTS

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Page 2, Line 3: Do the authors mean to use the indefinite article “a” instead of the definite article “the” in front of New European Driving Cycle?

Page 3, line 8: The sentence starting “In addition to . . .” is very long. The clarity of this sentence could be increased by splitting it into two sentences.

Page 3, Line 9: Do the authors mean the plural “processes” instead of the singular “process?”

Page 3, Line 11: There needs to be a “,” after distribution.

Page 3, Line 11: What do authors mean here by “different processes?” Are they referring to primary vs delayed primary aerosol?

Page 3, Line 13: For clarity, the authors should describe how secondary emissions differ from delayed primary. This has not been described in the manuscript yet.

Page 3, Line 18: Are there references for the previous studies using flow through chambers? That would help put this work into a better context for the reader.

Page 3, Line 20: There needs to be a “,” after both instances of “chamber”. Also, do the authors mean “Potential Aerosol Mass?” or “potential aerosol mass?”

Page 3, Line 23: Do the authors mean “secondary particulate emissions” or “Secondary Particulate Emissions.”

Page 4, Line 1: Here, the authors have defined secondary processes, but they have mentioned it several times already. This statement would be the most effective after the first mention of secondary particulate matter.

Page 4, Line 5: The acronym FVV has not yet been defined.

Page 4, Line 3: For the sentence starting “E.g.,” the use of “for example” in this sentence is redundant because E.g., is Latin for “for example.”

Page 4, line 8: Since BTEX is being defined, the authors do not need to put “BTEX

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=” in the parentheses. Furthermore, the right hand parentheses for this definition has been omitted.

Page 4, Line 15: It is not clear how the last two sentences of this paragraph corroborate the statement that “different photo-oxidation pathways are also dependent on conditions.” I would suggest either to delete these statements or add an additional statement that clarifies why this is important.

Page 4, Line 16: Do the authors mean to add the definite article “The” in front of “European union?”

Page 4, line 23: The authors should either place a “,” before typically, or move it towards the end of the sentence to read “hydrocarbon emissions are typically lower.”

Page 5, Line 6: The acronym PAM has already been defined.

Page 5, Line 11: Do the authors mean “focused on” instead of “focused to?”

Page 5, line 15: “First” after the semicolon does not need to be capitalized.

Page 5, line 15: I am confused what the urban driving cycle is as opposed to the NEDC, and how it is repeated twice if doing a cold start on a separate day is required.

Page 5, line 15: This sentence is missing many definite articles. It should read “the urban,” “the first,” “the second,” and “the last.”

Page 6, Line 14: “-3” in “cm-3” should be a superscript.

Page 7, Line 1: What metals and elements specifically?

Page 7, Line 1: The phrase starting “and DeCarlo” is not an independent clause. Perhaps deleted the “;” and add a “,” after “(2006)?”

Page 7, Line 31: The sentence starting “Total hydro carbons . . .” is entirely redundant because an almost exact replicate of this phrase was stated on Page 7, Line 27.

Page 7, Line 34: The sentence starting “In these measurements . . .” and the sentence
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that follows it on Page 8, Line 1 seems entirely out of place here. First, FTIR and HPLC measurements have not been mentioned yet. This sentence makes more sense if placed after the paragraph that runs from Page 8, Line 5 to Line 10. Finally, I believe the phrase “GC and HC” should be replaced by GC, FTIR, and HPLC” for this sentence to make sense.

Page 8, Line 13: Delete “-“ before “ratios.”

Page 9, Line 24: Please enclose “2015” in parentheses.

Page 9, Line 26: Please enclose “2015” in parentheses.

Page 9, Line 26: “O:C ratio” is a redundant phrase as the : indicates that it is a ratio. Please remove “ratio” in this instance and all instances thereafter.

Page 12, Line 27: Do the authors mean “studied the exhaust of secondary?”

Page 13, Line 13: There should be a “,” after “Thus.”

Page 14, Line 4: The phrase “for E10 largest” should read “for E10, the largest.”

Page 14, Line 8: The definite article “the” should come before “largest.”

Page 14, Line 9: The indefinite article “A” should come before “moderate.” Furthermore, the plus “were” should be changed to “was.”

Page 14, Line 15: The sentence that starts “For E10 . . .” is missing commas and definite and indefinite articles. It should read “For E10, the cold start had a dominating role in secondary aerosol formation, with a clear increase after the cold start . . .”

Page 14, Line 33: What does “ions” refer to in this case? Inorganic species, ions in the mass spectrum, or both?

Page 15, Line 6: Do the authors mean “dependent” instead of “depending?”