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From: est@ce.gatech.edu

Date: Thu, 15 Dec 2011 10:46:10 -0500

Subject: Revision requested for ES&T manuscript es-2011-03747f

To: dbs36@cornell.edu

Cc: est@ce.gatech.edu

15-Dec-2011

Journal: Environmental Science & Technology

Manuscript ID: es-2011-03747f

Title: "Contribution of Lubricating Oil to Particulate Matter Emissions from Light-duty Gasoline Vehicles in Kansas City"

Author(s): Sonntag, Darrell; Nam, Ed; Bailey, Chad; Fulper, Carl; Baldauf, Richard

Revision due before: 19-Jan-2012

Dear Dr. Sonntag:

I have now read through both the manuscript as well as the reviews of the article you recently resubmitted to Environmental Science & Technology. As you will note, the reviews are mixed. One major issue that is brought up is the novelty of the data and what sets this study apart from prior studies. Presentation is also cited. Given the various issues raised, I do not believe that the manuscript is suitable for publication in Environmental Science and Technology in its present form. A revised paper would have to carefully address the various comments made by the reviewers, making the appropriate revisions or giving reasons why the suggestions are not being followed. In conducting your revisions, please make sure that the final manuscript is less than 7000 word equivalents. It will be re-reviewed and the outcome is in doubt.

Should you decide to revise your manuscript, you should retransmit your files as a revision on the ES&T Web Submission page. Please include a detailed, point-by-point list of your replies to the comments of the reviewers as an attachment to your cover letter. For each comment, make a clear connection from it to your response so as to facilitate my simultaneous consideration of the reviewer's comment and your reply. Please show how the manuscript has been clarified. In addition, please note the important preparative and formatting

requests required for publication in ES&T.

I would like to thank you for considering ES&T for publication of your manuscript. If you have any questions, feel free to contact me.

On the basis of the reviewers' comments, please revise the manuscript considering all suggestions carefully, and either change the manuscript appropriately or provide convincing reasons for declining to do so. Please prepare a detailed, point-by-point list of your replies to the comments of the reviewers. For each reviewer comment, make a clear connection to your response, so as to facilitate my simultaneous consideration of the reviewers' comments and your replies to those comments. Please explain how the text of the manuscript itself has been clarified.

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Length: Please note that your manuscript's current word count is approximately 6670. ES&T guidelines indicate that an Article should be no more than 7000 word-equivalents. In brief, count text beginning with the abstract. Do not count the references or the figure and table captions. Add at least 300 words for each figure, scheme, or table. See the Instructions to Authors for details on how the word count is derived. Please make good use of Supporting Information by moving material of interest mainly to specialists into this supplementary documentation, if you have not already done so. Publication of your paper depends in part on your ability to keep your revised manuscript at or below the length limit. If this is not possible, it should not exceed its current word count. If the revision word count does exceed the current word count, you must provide a detailed and compelling explanation of your efforts in your cover letter.

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We are making a concerted effort to reduce our processing time from receipt to acceptance. To avoid inactivation of your manuscript, please return your revision by the due date. The revision of your manuscript is due before 19-Jan-2012. If you do not intend to revise the manuscript please let me know so that I can inactivate the

submission.

We look forward to considering your revised paper. Please contact our office if you have any questions or concerns as you proceed. Your interest in publishing your work in ES&T is appreciated.

Sincerely,

Armistead G. Russell
Associate Editor
Environmental Science & Technology
Phone: 404-385-6280
Fax: 404-894-8266
Email: EST@ce.gatech.edu

Reviewer(s)' Comments to Author:

Reviewer: 1

Recommendation: Accept, with minor revisions noted.

Comments:

The paper reports on useful work that certainly is of importance. Personally, the presentation is a bit "dry" and could be more entertaining. A few details passed my mind while reading, these should be considered / addressed in a minor revision:

- 1 - Trucks enter the discussion via the tables (Table 1) and supplementary material: please define Truck versus Car.
- 2- Besides numbers of vehicles (bottom of page 3), total cylinder volume or total driven miles may be a better reference to compare LDGVs versus diesel vehicles. The latter usually make more milage!
- 3- page 2 abstract line 6: the reader immediately wonders: 25% and 47%, where is what is the remaining 28% that constitutes PM - is it inorganics/metallics?
- 4- p4 explain SVOC (is now done on page 6)
- 5- p5 bottom : ... without changing..... Is it considered / accounted for that older vehicles, presumably owned by less wealthy people, drive on lower quality gasoline (cheaper pumps selling slightly off-season fuels)
- 6 - p6: a PM2.5 cyclone: OK, but please describe also the filter system in more detail. PM1.0 or PM0.1? What type, cut size etc.

- 7- p6 EC, OC, trace elements and ions: 1) why not smoke/FSN, and 2) where is the reporting on trace elements?
8. p7 equation1: how are intercept b and error e separated
- 9 more discussion on the intercept, which is quite important, would be in place. What is it and why?
- 10 p8 line 7: season: see my comment 5 above: fuel quality changes with season. Is that considered?
11. p 10 top: so (please confirm): X1 = Hopanes&steranes/organic carbon and X2= PAH/EC? Otherwise - be more clear
- 12 p 17 PM line 2: For the 1991-2004 ... PM. Is there an explanation for this, adding to the text that follows? Assessment is one thing, analysis is another and the paper is a bit lean on analysis & what the impact of it all is or should be.
- 13- where is ref 8 "submitted"? info missing.
- 14 - please check if all Supplementary info is really needed. Seems a bit excessive (compared to what is usually added as Supplementary info)

Additional Questions:

Rate the overall importance of this paper to the field of environmental science and technology (10 - High Importance / 1 - Low Importance): 7

Rate the originality/novelty of the submission (10 - High Originality / 1 - Low Originality): 7

Rate the technical quality of the submission (10 - Excellent Technical Quality / 1 - Poor Technical Quality): 8

Rate the clarity of the submission's presentation (10 - Excellent Presentation / 1 - Poor Presentation): 6

Reviewer: 2

Recommendation: Accept, with major revisions noted.

Comments:

Review attached

Additional Questions:

Rate the overall importance of this paper to the field of environmental science and technology (10 - High Importance / 1 - Low Importance): 8

Rate the originality/novelty of the submission (10 - High Originality / 1 - Low Originality): 6

Rate the technical quality of the submission (10 - Excellent Technical Quality / 1 - Poor Technical Quality): 6

Rate the clarity of the submission's presentation (10 - Excellent Presentation / 1 - Poor Presentation): 5

Reviewer: 3

Recommendation: Reconsider after major revisions; outcome in doubt.

Comments:
(Overall)

In this study, PM mass and their organic markers emitted from in-use 99 gasoline light-duty vehicles were measured based on the chassis dynamometer tests. Based on these experimental data with vehicle-population in the Kansas City, contributions of lubricating oil and gasoline fuel to the PM mass were estimated. Although this information (e.g. Fig.1 and Table 4) is valuable, the originality and importance of the method and data are not very high, and method/result are not well written. Furthermore, the origin of large proportions (about 10-30%) of the PM mass was undetermined. Therefore, I think this manuscript is not suitable for publication in ES&T in this present form. My concerns are listed below.

(Major concerns)

1. It should attribute the large intercept (undetermined proportion of PM, TC, EC, and OC) to fuel, oil, and etc. in some way. At least, the author should clearly explain the reason of the large intercept. If it is due to the inclusion of many vehicles in a simultaneous regression analysis, it may be better that each regression analysis is performed for a single vehicle, and the average and the range of the estimated

source contributions are shown.

2. It is unclear that what kind of data was used in the regression analysis, and how the analysis was conducted, and validity of the results. This is probably because of the writing of the text and shortness of information for calculation and analysis. For example, it is difficult to understand the meanings and grounds for the explanations at P5, L19-22 "Fifty-two chemical samples...". I recommend that author provide a figure in the manuscript that evokes image of the method and result of the regression analysis. Are the Figures S6 and S7 the results of the regression analysis? The explanations for these figures are quite insufficient.

3. It is better to discuss the GC/MS chromatogram patterns (e.g. Fig.1 of Brandenberger et al., 2005, and Fig.7 of Fushimi et al., 2011) of PM samples, fuel, and oil. This comparison indicate the origin of OC.

4. Please clearly state the reason why vehicles were split by 1991. It may be useful that source apportionments are conducted by the four (or more) categories shown in Table 1.

5. Please discuss the reason why oil-derived EC was very low for gasoline vehicles compared to diesel vehicles.

6. P16, L26-48: Many hopane and sterane compounds were measured in this study. Is it difficult to estimate the gaseous adsorption and evaporation using the measured profile data?

(Minor concerns)

1. Please show the survey year for the vehicle populations in the body text and the abstract.

2. Please describe in the abstract that chassis dynamometer tests were conducted, and the number of vehicles tested.

3. p3, L24: Is the name "Oil burners" reasonable? How about "High-oil-emitters"?

4. P4, L1: The words "fitting species" may be better to substitute to "marker species".

5. P5, L19: What the words “chemical sample” mean? Does it mean “the sample for chemical analysis”?
6. P6, L33-38: What the sentences “The composites ...” mean?”
7. P7, L2: What the words “phase measurements” mean?
8. Table 1: Please show in the table that what 100% for the vehicle populations is. Is the remaining 50% diesel vehicles? What the words “Sample Weights” mean?
9. P10, L10-11: The resolution of the words such as “Hopanes and Steranes/Organic Carbon” is too low.
10. Figure 1: Does the “Fleet” mean the whole vehicles? If so, I think the figure of the Fleet should be omitted. Please change the order of the legend corresponding to the figure.
11. Table 3: Title of the second column “Model” should be a mistake. “Component” or something?
12. P8, L20; P16, L54: The words “polycyclic aromatic hydrocarbons (PAH)” were shown 3 times.
13. Supporting Information: Units are not provided (e.g., Figs S2 and S3). Figs. S4 and S5 are not quoted in the text.

(References)

Brandenberger, S., Mohr, M., Grob, K., Neukom, H.P., 2005. Contribution of unburned lubricating oil and diesel fuel to particulate emission from passenger cars. *Atmospheric Environment* 39, 6985-6994.

Fushimi A., Saitoh K., Fujitani Y., Hasegawa S., Takahashi K., Tanabe K., Kobayashi S. 2011. Organic-rich nanoparticles (diameter: 10–30 nm) in diesel exhaust: Fuel and oil contribution based on chemical composition, *Atmospheric Environment*, 45, 6326–6336.

Additional Questions:

Rate the overall importance of this paper to the field of environmental science and technology (10 - High Importance / 1 - Low

Importance): 5

Rate the originality/novelty of the submission (10 - High Originality / 1 - Low Originality): 5

Rate the technical quality of the submission (10 - Excellent Technical Quality / 1 - Poor Technical Quality): 4

Rate the clarity of the submission's presentation (10 - Excellent Presentation / 1 - Poor Presentation): 4

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