Responses to Comments

I. Responses to the comments from Reviewer 1

Comment 1

The paper reports on useful work that certainly is of importance. Personally, the presentation is a bit "dry" and could be more entertaining.

Authors' Response: Substantial revisions are done to improve the readability of the paper, and to provide additional justifications for the work conducted. The introduction is re-written to emphasize the importance of the work. Additionally, clearer explanation of the methods used, comparison of the data used with the complete KCVES data (Figure 1) should make the paper easier and more interesting to read. Also, the discussion added on the limitations/uncertainties (page 17-19) provides greater context where the results can be used. We believe these changes make the results more accessible and interesting for the ES&T reader.

Comment 2

Trucks enter the discussion via the tables (Table 1) and supplementary material: please define Truck versus Car.

Authors' Response: We appreciate the reviewer pointing out this oversight. A footnote is added to Table 1 which includes the following definition: "The definition for Car and Truck is taken from the 2004 Kansas City Travel Behavior Survey (22) Passenger cars are defined as coups, sedans, and wagons, trucks are defined as minvans, sport-utility vehicles, and pickups."

Comment 3

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 mileage!

Authors' Response: The discussion in the introduction on comparing diesel vehicle PM and LDGV PM has been removed. The Introduction is now more focused specifically on the lubricant contributions to LDGV. Discussion is added on the contribution of high-oil emitting LDGVs compared to normal LDGVs. (Paragraph 3 on page 3).

Comment 4

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?

Authors' Response: *Because the main focus of the paper is on the lubricating oil contribution to PM, only the lubrication oil contribution is mentioned (25%). The abstract now mentions that :*

"The PM is not completely apportioned to the gasoline and oil due to several contributing factors, including: varied chemical composition of PM among vehicles, metal emissions, and PM measurement artifacts. Additional uncertainties include potential sorption of polycyclic aromatic hydrocarbons into the oil, contributions of semi-volatile organic compounds from the oil to the PM measurements, and representing the in-use fleet with a limited number of vehicles."

The contribution of inorganics/metallic are discussed in the Results in the Gravimetric Mass subsection (page 17), as well as the third point in the Uncertainty and Limitations Section.

Comment 5

p4 explain SVOC (is now done on page 6)

Authors' Response: *SVOC is now spelled out on page 3, at the first mention of SVOCs:*

" Lubricating oil (also known as engine oil, motor oil, and engine lubricant) coats the piston and cylinder walls, from which semivolatile organic compounds (SVOC) desorb during the exhaust stroke, providing a major pathway for oil consumption and PM emissions for properly functioning LDGVs. SVOCs are higher molecular weight hydrocarbons that partition between the gaseous and particle phases. Some SVOCs nucleate or condense onto particles as exhaust dilutes (*8*)."

Comment 6

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)

Authors' Response: *We agree with Reviewer 1 that fuel properties can vary substantially. Fuel property variability is certainly a source of variability in the PM emission rates. Fuel samples were analyzed on a subset of vehicles in the KCVES reported in EPA (2008), which did show substantial variability in sulfur levels. The impact of income distribution on fuel properties would require knowing the income of the owner associated with each vehicle, associating private income data with each vehicle. Conducting such analysis was beyond the design and scope of this project.*

The following sentence is added to the uncertainties/limitations of the study regarding the impact of different fuel properties on page 19:

"6. The relative oil contribution to PM emissions of the in-use vehicle fleet has likely changed since 2004 due to fleet-turnover, new vehicle technologies, changes in fuel properties, and implementation of the Tier 2 Vehicle & Gasoline Sulfur Program Final Rule (Tier-2). The average fuel sulfur content for a subsample of 40 vehicles in round 2 of the KCVES was 203 ppm, (26) far above the average fuel sulfur content of 30 ppm set by the Tier 2 (40)."

Comment 7

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.

Authors' Response: Additional experimental information is included in the Supporting Information: Figure S1, and Table S1. Table S1 includes detail and specification regarding the instruments and samplers used in the sampling program. Specifically, the description of the PM2.5 cyclone is provided:

"Bendix 240 cyclone, flow rate set at 113 lpm to set cutpoint at ~ 2.5 um; 100% penetration of particles smaller than 1 um (Molenar, 1997)"

Comment 8

p6 EC, OC, trace elements and ions: 1) why not smoke/FSN, and 2) where is the reporting on trace elements?

Authors' Response: 1) EPA has never measured visible smoke from LDGV and discontinued its regulations/measurements on visible smoke form HDDV once PM emission standards were in place. The correlation between visible smoke and PM mass was only qualitative. Also, it is unclear how smoke/FSN can be related to ambient PM mass concentrations.

2)The experimental information includes discussion on the protocol used to measure trace elements. Discussion of results of trace elements is updated in the draft, and included in on page 17, in discussing the contribution of inorganic elements, and metals to total PM emission rates. The emission rates for each element are presented in Table S5.

Comment 9

p7 equation1: how are intercept b and error e separated. More discussion on the intercept, which is quite important, would be in place. What is it and why?

Authors' Response: Additional discussion is included on the intercept term and the model residuals. The model residuals are discussed in the Model Diagnostics subsection regarding

Additional discussion is given on the intercept as well. This comment is responded to in more depth from Reviewer 3, comment 2.

Comment 10

p8 line 7: season: see my comment 6 above: fuel quality changes with season. Is that considered?

Authors' Response: We agree with the reviewer that fuel properties change with season. For a subset of vehicles that were analyzed for the fuel properties, the summer vehicles had noticeably lower Reid Vapor Pressure and higher Aromatic content, than the fuel sampled in the winter.

The regression model does not take into account fuel properties. It assumes that the relative contribution of PM per ug/mile of PAH fuel markers is the same across season. However, the model treats the winter and summer equally, to correct for any unbalance in the study design (i.e. more vehicles tested in winter, compared to summer). Figure S4 and S5 labels the observations separately by season. Fitting separate models by season was explored. However, these models still had high intercept terms, and the significance of the results decreased substantially. Thus, the model was fit to the observations from both seasons pooled together. Thus, no changes were made to the manuscript. This discussion is included in the Methods Section, under the subsection entitled: 'Model Coefficients' on page 12:

"The large intercept for the pre-1991 vehicles is attributed to the slight increase in the $\frac{\text{Hopanes and Steranes}}{\text{Organic Carbon}}$ ratio (Figure S4) within the samples in the group. The samples were split into smaller groups (by season, pre-1981 and 1981-1990 model year groups, and truck vs. car) in an attempt to improve the homogeneity of the particulate to marker ratios in a way that the intercept was no longer significant to the model predictions. When the data was fit to smaller groups, the intercept retained its high significance, and the model coefficients for the gasoline and oil markers became largely dependent one or two observations."

The impact of fuel properties is considered discussed in the uncertainty limitations and discussion on page 19:

Comment 11

p 10 top: so (please confirm): X1 = Hopanes&steranes/organic carbon and X2= PAH/EC? Otherwise - be more clear

Authors' Response: The paragraph on the Survey Regression Model is revised (page 9).For example, y is replaced by PM in the regression model terminology. X1 and X2 are clarified by being explicitly defined:

$PM = b + m_1 x_1 + m_2 x_2 + e$ (1) were *PM* is the elemental carbon (EC), organic carbon (OC), total carbon (TC), or Teflon-gravimetric mass (MASS) emissions; *b* is the intercept, *x_1 is the fuel markers emissions (ug/mi); and x_2 is the oil markers emissions (ug/mi)*; m_1 is the slope of the fuel-associated emissions; m_2 is the slope of oil-associated emissions; *e* is the residual error.

Comment 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.

Authors' Response: This paragraph is removed from the revised draft. The impact of the result of larger standard errors was deemed of insufficient impact to be discussed in the results section.

We appreciate the reviewer's comment on providing analysis of the impacts of the study. The subsection, "Uncertainties and Limitations" was added, in part, to provide better synthesis and analysis of the results, by placing the results in greater context, and discussing areas where more research is needed.

Comment 13

where is ref 8 "submitted"? info missing.

Authors' Response: The Fujita, Zielinska, Chow et al. reference has been submitted to the Journal of Air & Waste Management Association. However, since the journal has not yet formally accepted the manuscript, it has not been added. If the paper is still not accepted at the time of publication of this draft, the reference of the Final Report of the study can be provided, which supports the citations made in the manuscript. "COLLABORATIVE LUBRICATING OIL STUDY ON EMISSIONS (CLOSE)"

Comment 14

please check if all Supplementary info is really needed. Seems a bit excessive (compared to what is usually added as Supplementary info)

Authors' Response: The Supplemental Information has been shortened. Only Figures and Tables that are referenced in the main text are included.

II. Responses to the comments from Reviewer 2

Comment 1

Fundamentally, there should be better presentation of structure and results. For instance, 'Kansas City Light-Duty Vehicle Emissions Study' should be part of method section….The text tries to be too much of KCVES instead of a clear and well developed story line.

Authors' Response: We appreciated the Reviewer's suggestion to improve the readability and flow of the Methods section. We revised the order and the content of the Methods section to include the following subheadings: Data, Survey Regression Model, Gasoline Fuel Markers, Lubricating Oil Markers, Application to the Kansas City Light-Duty Gasoline Fleet, Model Diagnostics, and Model Coefficients.

The subsection ' Data' is re-written to emphasize the aspects of KCVES that are pertinent to the current study. We believe that the 'Methods' section now stands on its own better, and sets up the reader to better understand, and focus on the Results Section.

Comment 2

One of the main issues with the presentation is that there is insufficient description of hopanes, steranes, and PAHs analysis –Which method was used for the analysis? A Standard one or an in house method?

Experimental Methods – line 19-39 in page 6, as mentioned above, the author should provide detail methods of PAHs and SVOC analysis.

Authors' Response: Additional information on the experimental methods and organic speciation analysis is provided in the supporting information. These are referred to in the main text. In Table S1, the description is added of the Gas Chromotograph/Mass Spectrometer:

"Varian 1200 triple quadruple gas chromatograph/mass spectrometer (GC/MS/MS) system, or Varian coupled to a Saturn 2000 ion trap mass spectrometer system with MS/MS and chemical ionization capabilities."

And Table S2:

"In addition, the GC/MS measured 95 semi-volatile and particulate PAH, 49 alkanes, 99 polar organic compounds, and 25 nitro-PAHs. Method detection limits are 0.01-0.03 ng/µl for PAH, hopanes and steranes. Details on DRI's laboratory analytical methods, including deuterated internal standards are given in EPA (2008)."

Comment 3

There are assumptions for 'multiple regression analysis'-how were correlations between fuel/oil markers and EC, OC, TC and PM?

Results and discussion - What are the assumptions for multiple regression analysis used in this study? e.g. were the variables correlated? were they normal distributed?

Authors' Response: As the model estimates are the main contribution of the paper, we agree that it is important to present the model assumption and model diagnostic evaluation of these assumptions in the main text. We added the subsection "Model Diagnostics" on page 11.

The section evaluates the assumptions of normal, independently-distributed residuals with mean of zero. Through the text, and refers to two additions in the Supporting Information, Table S5. Standard Normality Tests of Survey Regression Residuals, and Figures S6-S7. Residual Plots.

We agree with the reviewer, that correlation of predictive variables (multicolllinearity) can impact the results of linear models. Multicollinearity inflates the standard errors of the predictors, causing wide swings in estimates in model coefficients, and can cause unreasonable relationships to be estimated with coefficients.

The correlation of the two variables is evaluated. The R2 between the oil and gasoline markers is 0.21 (for the 1991-2004 vehicles) and 0.38 for the pre-1991 vehicles. This relates to a variance inflation factor (VIF) of 1.27 and 1.62. Thus, the variances of the coefficients are slightly inflated, but the correlation is not cause of alarm. (VIF > 10 would be a cause for alarm). Also, the predictive variables are generally highly-significant in the regression models (Table 2). Due to lack of space, and limited conclusions from the multicollinearity analysis, the impact of multicollinearity is not discussed in the paper. Instead, the paper focuses on the model diagnostics of the residuals, and the applicability of the linear assumptions and the resulting intercept term in the 'Mode Coefficients' Section.

Comment 4

Introduction – Line 12-20 in page 4, 'In addition to……(16,17,18)' the author should extend the discussion in the paragraph.

Authors' Response: The paragraph on health impacts of lubricating oil emissions is expanded to include additional discussion of the contribution of SVOCs and ultrafine particles. The discussion is included on Page 3:

Comment 5

Line 21-50 in page 4, the author may focus on findings of previous studies, such as development methods and validating markers for apportioning PM emissions.

Authors' Response: The paragraph is revised to focus on markers measured from emission characterization studies and receptor modeling studies.

The discussion on the results of Kleeman et al. and Fujita et al. contributions are moved to the Methods section to consolidate the discussion of the methods section.

Comment 6

Line 36 in page 6, 'the compositing reduced the 102 individual vehicle tests to 26 individual……' how were those 26 selected? Were they typical? How many tests for each vehicle type and age group? 26 for both PAHs and SVOC or for each?

Authors' Response: Substantial effort was made to clarify the data used in conducting the analysis, and in evaluating how representative the samples were. The data section was re-written to emphasize that the data chemical samples were conducted on 102 tests obtained from a subset of the entire KCVES study. Two sentences was added/modified to clarify how the composites were made. (Page 6)

"Although, the composite chemical samples contained PM emissions from more than one vehicle, the vehicles that comprised each composite sample were within the same strata, and had similar BC/PM ratios (21). The compositing reduced the 102 individual vehicle tests selected for chemical analysis to 52 individual and composite chemical samples (26 in each season) as shown in Table 1 (21)."

A new paragraph on page 7-8, Figure 1, Figure S2, and Figure S3, were added to evaluate how the vehicles selected for chemical analysis were representative (typical) of the randomly sampled KCVES vehicles.

Comment 7

Section 'Elemental Carbon' and 'Organic Carbon' - there is not sufficient discussion of the results in terms of comparisons with other studies.

Authors' Response: The results from our study are compared to results obtained by Kleeman et al. (2008) and Schauer et al. (2002). Unfortunately, little research has been conducted on lubricant's contribution to EC and OC emissions from LDGVs.

Comment 8

figures in SI could be moved into the main text…
Presentation – images can demonstrate results more efficiently, hence, figures in SI could be moved into the main text

Authors' Response: Unfortunately, moving all the figures into the main text would cause the manuscript to be too long. The Figures that are in the main text (Figures 1 and 2) are important for the every-day reader to understand the representativeness of the vehicles selected in the study, and to understand the results. The figures left in the supporting information are more concentrated on additional experimental information, model diagnostics, model assumptions, and are left in the supporting information for the interested reader.

Comment 9

Section ‘Estimate from Continuous PM Emission Data’ in page 18 – is this another method of estimation? It hasn’t been mentioned in the early sections, especially the section of method. What’s the relation between these two approaches? Photoacoustic instrument is based on optical method to measure BC concentrations. I’ve seen a lot of studies on the comparison of optical methods and IMPROVE for BC measurements, which indicated that big differences between these two methods. Then, how can these two estimates be evaluated?

<u>Authors’ Response:</u> This section was removed from the text. It was included in the previous draft to provide a secondary estimate of the lubricating oil contribution from the KCVES. However, the strength of the paper is based on the survey regression estimates, and the inclusion of the secondary approach seemed to detract from the clarity and strength of the paper. As such, the paper focuses on the survey regression estimates, which enables more space to clearly discuss the data, methods, results, and discussion of limitations and uncertainties of the study.

III. Responses to the comments from Reviewer 3

Comment 1

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.

Authors' Response: We appreciate the reviewer's honest opinion. Much of the revisions were focused on these two comments: provide clarity and motivation behind the novelty of the results, and improve the presentation of the methods and results. These major revisions taken in response to these comments are documented in the cover letter.

Comment 2

Furthermore, the origin of large proportions (about 10-30%) of the PM mass was undetermined….

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.

Authors' Response: Additional Discussion on the rationale for including the intercept is included in the paper. On page 12, the additional sentence is added to the Model Coefficients section.

"Unlike Kleeman et al. (15) and Fujita et al (16), the intercept is retained in the model to enable unbiased estimates of the mean emission rates, prevent misfits of the model at low PM levels, and to avoid placing strong assumptions on the data."

A paragraph is added in the Uncertainty and Limitations section : (page 18)

"3. Inclusion of the intercept likely causes the contribution of the fuel and lubricating oil to be underestimated. The metal wear emissions and positive measurement artifact are physical rationale behind a portion of the intercept. However, these are unable to explain much of the intercept, particularly for the pre-1991 vehicles, which had an intercept of ~13 mg/mi. As explained earlier, the intercept is an outcome of fitting a pooled data set by a relatively simple linear model. Thus, some of the intercept emissions are in reality contributed by the lubricant and/or the fuel, however the contribution

attributable to each source is unknown. Even though the intercept prevents the complete allocation of PM to the fuel and lubricating oil, the intercept is retained to maintain transparency regarding the uncertainties and limitations involved in the apportionment of the PM emissions."

We agree with the reviewer that it would be better to perform the regression analysis on smaller groups, and individual vehicles if possible. However, we only have one observation per chemical sample. We revised the discussion on the methods section to make this clearer. Discussion is included in the Model Coefficients section that discusses why the regression models were not fit to smaller groups of data:

"When the data was fit to smaller groups, the intercept retained its high significance, and the model coefficients for the gasoline and oil markers became largely dependent one or two observations. Thus, the intercept is retained in the model as an additional source of uncertainty, introduced from approximating a relatively simple linear model to the complex relationship between markers and particulate emissions."

Comment 3

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.

Authors' Response: As discussed above in the Editor's comment, the Methods section was revised to clarify the data used in the regression analysis.

We appreciated the Reviewer's comment to include a Figure that clarifies the data and method used in the regression analysis. This comment sponsored the inclusion of Figure 1 in the paper (page 8), which is intended to both clarify what data were used in the regression analysis, and to examine how representative the data are.

Comment 4

Are the Figures S6 and S7 the results of the regression analysis? The explanations for these figures are quite insufficient.

Authors' Response: Previous Figures S6 and S7 are not referred to in the main text, and have been removed from the Supporting Information.

Comment 5

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.

Authors' Response: We appreciate the Reviewer bringing these articles to our attention. We are familiar with using chromatogram patterns, particularly from GC-FID measurements to calculate the contribution of lubricant oil, but this approach was not taken for this project for several reasons. Brandenberger et al. 2005 used GC-FID measurements to calculate the contribution of lubricant to paraffins.This is useful because the carbon distribution can be obtained from the GC-FID measurements. However, the KCVES did not collect GC-FID measurements, needed to perform such an analysis. Additionally, the scope of our project was also to calculate to the contribution of lubricant to the gravimetric mass measurements, not just to the organic particulate.

We also agree with the Reviewer that looking at mass chromatogram patterns from GC-MS is useful to first qualitatively examine the potential lubricant contributions to organic particles, as was done in Fushimi et al. 2011. However, the patterns have their limitations, from mass spectrometry, the patterns are snapshots at (m/z) ratios, and cannot be used quantitatively calculate the lubricant contribution to PM. In fact, Fushimi et al. 2011, used a hopane tracer,17α(H),21β(H)-hopane, (similar to our project) to quantitatively calculate the lubricant oil contribution to the nanoparticle emissions. Because the objective of the project was to estimate a number—the lubricant contribution to PM in the in-use fleet, only the tracer method was pursued in the project.

As discussed in the Response to the Editor's comments, the manuscript was substantially revised to clarify the objective of the paper—to yield a quantitative estimate of the oil-to-PM contribution in the in-use fleet, with additional discussion in the Introduction and the Results Section. This was done to clarify why we used the survey regression/tracer method. Due to shortness of space, the alternative approaches of using gas chromatograms are not discussed in the main text.

We appreciate the Reviewer bringing these articles to our attention. The Brandenberger et al. 2005 is now referenced in the "Uncertainty and Limitations" Section under the impact of operating conditions, (page 19). Also, Fushimi et al. 2007, was included in the introduction on the discussion of lubricating oil contribution to ultrafine particles.

Comment 6

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.

Authors' Response:

The reason the vehicles were split according to the sampling design strata has been clarified in the text. In the abstract:

"Vehicles are analyzed according to pre-1991 and 1991-2004 groups due to differences in properties of the fitting species between newer and older vehicles, and to account for the sampling design of the study."

In the Methods section (page 11):

"A survey regression model is implemented by incorporating the statistical weights as presented in Table 1 into the regression estimates. The sample weights are calculated as the ratio of the vehicle strata population to the number of chemical samples in the strata. The statistical weighting corrects for the oversampling of older vehicles and trucks in the KCVES compared to their population in the KC fleet."

Additionally, it was attempted to calculate the lubricating oil contribution to PM separately for each of the four model year group. However, it did not improve the model estimates, and reduced the significance of the important variables. Thus, it was decided to group the data into two model year groups, as discussed in page 10:

"The samples are grouped into two groups: pre-1991 and 1991-2004 because these groups have relatively constant marker to emission ratios within each group, while being large enough to estimate statistically significant relationships."

And on page 12:

"The samples were split into smaller groups (by season, pre-1981 and 1981-1990 model year groups, and truck vs. car) in an attempt to improve the homogeneity of the particulate to marker ratios in a way that the intercept was no longer significant to the model predictions. When the data was fit to smaller groups, the intercept retained its high significance, and the model coefficients for the gasoline and oil markers became largely dependent one or two observations."

Comment 7

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

Authors' Response: The scope of the paper is focused on LDGV PM emissions. Low oil-derived EC emissions correspond to results from the work by Kleeman et al. (2008) and Fujita et al. (2012). Kleeman et al. (2008) does estimate significant oil-to EC contributions in some of the Heavy-Duty Diesel Vehicles in their study, which they surmise could be an effect of different engine loads. However, our study did not shed any light on this issue, so it is not discussed in the results.

Comment 8

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?

Authors' Response: Hopanes and steranes are now described as ~C27-C45 compounds, which are relatively nonvolatile. In ambient air, they are virtually nonvolatile. No previous studies we have reviewed employing dilution sampling of the type used for measurement of LDGV emissions have reported these oil biomarkers in vapor or gas phase. For example, Schauer, J.J.; Kleeman, M.J.; Cass, G.R.; Simoneit, B.R.T. Measurement of Emissions from Air Pollution Sources. 5. C1-C32 Organic Compounds from Gasoline-Powered Motor Vehicles. Environ. Sci. Technol. 2002, 36, 1169-1180.

The high molecular weights of these compounds make them extremely non-volatile. For example, hopane (17α(H),21ß(H)-hopane) has a molecular weight of 412.7, a boiling point of 457 ˚C at 760 mmHg, and a vapor pressure of 0 mmHg at 25˚C (as reported by chemspider.com) .

Comment 9

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

Authors' Response: The dates of the study are now included in the abstract and the Data section:

Abstract:

"The contribution of lubricating oil to particulate matter (PM) emissions is estimated from in-representative of the in-use 2004 light-duty use gasoline vehicles fleet is estimated from tested in the Kansas City Light-Duty Vehicle Emissions Study (KCVES)."

Page 4
*"*The KCVES tested 496 vehicles in two rounds. Round 1 (summer) ran from July – September 2004, and Round 2 (winter) from January – April 2005."

Additionally, the dates and source of the vehicle population, are now included in the footnote of Table 1.

The population of vehicles is certainty larger in 2004 than 2000. However, because we are estimating the mean PM emission rates, and because the sample is such a small percentage of the in-use fleet (population is needed for finite population correction in survey samples—but is not needed here), the exact number of vehicles on the results is inconsequential. The statistical weights are used in the survey regression model. Using statistical weights based on an approximation of the 2004 vehicle population is done to give the reader an understanding of the small number of samples taken in comparison to the number of vehicles in the in-use Kansas City fleet.

To emphasize this limitation of the study, the following paragraph is added in the 'Uncertainty and Limitations' section (page 18):

"4. Significant effort in both the KCVES study design, and the selection of the chemical samples was made to assure the samples are representative of the in-use fleet. Through the use of the JRR resampling method, the impact of excluding observations from the sample was quantified. Despite these efforts, the number of vehicles tested for chemical analysis are still limited, and uncertainties exist about the representation of the sample to both the Kansas City Metropolitan Area, and other areas of the United States. For example, the vehicles selected for chemical analysis appear to slightly under-represented by higher emitters in the newer vehicle strata (1996-2004), which could lead to an underestimation of the lubrication oil contribution."

Comment 10

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

Authors' Response: This information is included in the abstract in this sentence. We believe that explicitly most interested readers will implicitly understand the testing was conducted on a chassis dynamometer when we mention that the testing was conducted on LDGVS on the California Unified Driving Cycle.

"PM emissions are apportioned to lubricating oil and gasoline using aerosol-phase chemical markers measured in PM samples obtained from ninety-nine vehicles tested on the California Unified Driving Cycle."

The use of the chassis dynamometer is specifically mentioned on page 5, in the Data subsection:

"The vehicles were tested on a chassis dynamometer at ambient temperatures, in a garage with open bay doors, after soaking in the test facility overnight."

And the description of the chassis dynamometer is now included in the supporting information, Table S1.

Comment 8

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

Authors' Response: We agree with that the term "Oil burners" is too informal, and may be misleading. This is changed to "high-oil emitters", and "high-oil emitting vehicles."

Comment 9

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

Authors' Response: We believe both terms portray the intended meaning. Kleeman used 'tracer', Fujita et al. 2012 used 'Marker.' CMB studies use "fitting-species" which are often normalized by EC, OC, or TC concentrations. Because the terminology is kept consistent with Fujita et al. 2012, we have kept the use of "marker."

Comment 10

P5, L19: What the words “chemical sample” mean? Does it mean “the sample for chemical analysis”?

Authors’ Response: Similar response if given to Reviewer 2, Comment 7.

The “Data” subsection is re-written to emphasize the two stages of testing, which include the adding the sentences, and re-writing the subsequent paragraphs: (Page5)

“The data used to estimate the lubricating oil contribution to PM were collected from 102 vehicle tests (99 separate vehicles, with 3 repeat tests) that were selected for chemical analysis. The data was collected at two stages of testing, (1) PM measurements from each vehicle, and (2) Chemical analysis.”

Comment 11

P6, L33-38: What the sentences “The composites …” mean?”

Authors’ Response: Similar response if given to Reviewer 2, Comment 7. Similar response if given to Reviewer 2, Comment 7.

The text was revised on Page 6

“For the pre-1991 vehicles, the organic speciation was conducted on particulate obtained from one vehicle test. For the newer vehicles (1991-2004), the particulate from 2-5 vehicle tests was solvent extracted and composited together (depending on the amount of particulate emitted per vehicle) to assure sufficient extractable material for chemical analysis. Although, the composite chemical samples contained PM emissions from more than one vehicle, the vehicles that comprised each composite sample were within the same strata, and had similar BC/PM ratios (21). The compositing reduced the 102 individual vehicle tests selected for chemical analysis to 52 individual and composite chemical samples (26 in each season) as shown in Table 1 (21).”

Comment 12

P7, L2: What the words “phase measurements” mean?

Authors’ Response: The “Data” subsection is re-written to emphasize the two stages of testing, which include the adding the sentences, and re-writing the subsequent paragraphs: (Page5)

Regarding this comment, the phase-specific PM measurements from each vehicle were clarified on page 6

“In the first stage, the PM emission rates, including gravimetric mass, elemental carbon (EC), organic carbon (OC), and total carbon (TC), were sampled from each vehicle tested on the three-phase California Unified Driving Cycle, also known as the LA92 driving cycle. The LA92 includes a cold start (Phase 1), hot-running (Phase 2) and a hot-start (Phase 3), and of 9.8 miles of driving representative of arterial and freeway driving in Los Angeles, California.

Comment 13

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?

Authors' Response: Table 1 is revised. The column heading for (% of KC LDGV vehicle population) is changed to (% of KC LDGV vehicle population). The % are changed so that the LDGV strata in each season sum to 100% (rather than 50%). Definitions and data sources are provided in the footnotes.

Comment 14

P10, L10-11: The resolution of the words such as "Hopanes and Steranes/Organic Carbon" is too low.

Authors' Response: We will work with the manuscript editor to assure that the "Hopanes and Steranes/Organic Carbon" *and* "PAH/Elemental Carbon*" ratios have good resolution in the final text.*

Comment 15

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.

Authors' Response: As mentioned in the response to the Editor comment 1, the text is revised to emphasize that the major objective of the project is to estimate the representative oil-to-PM contribution from the in-use fleet. As such, we believe it is important to retain the "Fleet" estimated contribution in Figure 2.

Comment 16

Table 3: Title of the second column "Model" should be a mistake."Component" or something?

Authors' Response: It was referred to as 'Model' because separate regession models were fit to each measure of PM. It appears that this is confusing to the reader, as such Model (y) is changed to PM component, with four PM components: EC, OC, TC, and Gravimetric Mass (MASS).
These are explained in detail on page 9:

$y = b + m_1 x_1 + m_2 x_2 + e$ (1) were *PM* is the elemental carbon (EC), organic carbon (OC), total carbon (TC), or Teflon-gravimetric mass (MASS) emissions;"

Comment 17

P8, L20; P16, L54: The words "polycyclic aromatic hydrocarbons (PAH)" were shown 3 times.

Authors' Response: It is now only shown once at the first mention of PAH. (Page 8)

Comment 18

Supporting Information: Units are not provided (e.g., Figs S2 and S3). Figs. S4 and S5 are not quoted in the text.

<u>Authors' Response:</u> Missing Units are added to these figures (now labeled as Figure S4, S5, S6, and S7). S6 and S7 are now referred to in the text in the Model Diagnostics subsection (page 11).