Response to Referee 1:

Referee 1 raises two important concerns about our methods.

1. Contributions of LDV to PM Emissions
2. Goodness of fit for PM:CO enhancement ratios

Concern 1: Contributions of LDV to PM Emissions

Reviewer 1 is concerned about our assumption that HDVs are responsible for the bulk of $PM_{2.5}$ emissions, citing Habre et. al, 2020, which found LDVs responsible for a large percentage of $PM_{2.5}$ emissions in the LA region. Although this is an important concern, the methodology use (and consequently findings) and context examined by Habre et. al, 2020 differ enough from the methodology and context of this work, that we question the relevance of this paper's findings to our study.

Methodological Differences: Unlike the work we present, Habre et. al, 2020 does not use hourly filter measurements measuring total $PM_{2.5}$ mass, but instead uses filter measurements collected over full months (via two two-week samples) for their analysis. Habre et al., 2020 then use speciation data from filter samples in combination with traffic factors, to run the EPA Positive Matrix Factorization (EPA PMF v5.0) model. Using this model, Habre et al., 2020 estimate fraction of total aerosol burden attributable to various roadway sources for three size fractions, quasi-ultrafine (0.-.2 µm), fine (.2 - 2.5 µm), and coarse (2.5 -10 µm), and are able to explain 63%, 86%, and 88% of the variability in the quasi-ultrafine, fine, and coarse aerosol mass. Through this modeling Habre et al., 2020 estimate that LDV contribute 72% of tailpipe emissions in the fine size fraction by mass, and that abrasive vehicle emissions (not differentiated as HDV or LDV) make up 35% of the fine size fraction of traffic emissions by mass. However, these factors together make up only 32% of the explained fine aerosol mass and Habre et al., 2020 do not provide error estimates on these percentages. Furthermore, although Habre et al., 2020 quantifies the relative burden of LDV v. HDV tailpipe emissions, they do not attempt to calculate emission factors for either, making it difficult for us to compare their results to our own.

Contextual Differences: As opposed to our work, which focuses on spatially and temporally on highway emissions, Habre et al., 2020 analyzes aerosol measurements over wide areas, meaning that their analysis is meant to include emissions from surface streets, where vehicles tend to drive more slowly but brake more frequently. There are a number of reasons that we expect the relative contribution of LDV and HDV to aerosol mass to be different on highways v. surface streets. First, the percentage of vehicles that are HDV on surface streets is likely lower than what would be expected on highways. For example, in the SF Bay Area, EMFAC2017 estimates HDV fraction of all traffic driving at 30 mph to be ~0.7%, compared to ~4.4% for vehicles driving 60 mph. Furthermore, Habre et al., 2020 excludes data from the HDV-rich area of Long Beach in drawing conclusions related to the relative contributions to aerosol burden of HDV as compared to LDV. Because Habre et al., 2020 does not report the percentage of HDV in the area studied or attempt to quantify HDV or LDV emission factors, making a direct comparison difficult.

Beyond the contribution of HDV v. LDV to tailpipe aerosol emissions, Reviewer 1 is also concerned that a large fraction of highway PM emissions may be from abrasive vehicle emissions (AVE), including tire wear and brake wear. We do not dispute this idea, especially in recent years as HDV tailpipe controls have been put in place, substantially reducing emissions from HDV tailpipes, and we have clarified the text of our paper (L181-182) to make that more clear. Reviewer 1 is concerned that a large portion of AVE may be from LDV. However, on a per vehicle basis, HDV are expected to contribute significantly more to AVE. Estimates of the ratio of LDV and HDV AVE emission factors from previous studies vary widely and often consider PM_{10} or total mass loss (rather than $PM_{2.5}$) due to tire or brake wear. For example, MOVES3 Break and Tirewear Emissions (EPA, 2020) finds a wide range of total tire wear mass ratios per mile (HDV:LDV), ranging from 2.3 (Bauman et al., 1997) to 26 (Senco, 1999), with most estimates lying (Garben et al, 1997; Gebbe et al., 1997; EMPA, 2000) in the low to mid teens. Generally, tire wear is expected to increase with tire number and weight. HDV have ~4x more tires and ~15x the weight of LDV. The MOVES3 Brake and Tirewear Emissions (EPA, 2020) finds similarly large range of emission factor ratio estimates for brake wear (EPA, 2020). Only one HDV:LDV emission factor ratio was reported for $PM_{2.5}$. Abu-Allaban et al., 2003 reported a value of 3. However, for PM_{10}, the HDV:LDV emission factor ratio reported by previous literature ranges from 0.7 (Carbotech, 1999) to 24 (Rauterburg-Wulff, 1999) with most estimates (Luhana et al., 2004; Abu-Allaban et al., 2003; Westurland, 2001) in the range of 6-8.

Tailpipe emissions are similarly expected to be much higher on a per vehicle basis for HDV compared to LDV. In the studies we cite in Table 1 of the text, Geller et al., 2005 reported an HDV: LDV $PM_{2.5}$ emission rate ratio of 14, Ban-Weiss et al., 2008 reported a ratio of 20, and Li et al., 2014 reported a ratio of 7.5. Park et al., 2011 reported a lower ratio of 4.8, although we note that this was on surface streets rather than a highway. Similar to the studies we cite, at 60 mph, the EMFAC2017 model yields a HDV: LDV $PM_{2.5}$ emission rate ratio of 23 in 2009 and 8 in 2020.

Using literature-reported emission factor ratios, we put our assumption that HDV are responsible for the bulk of $PM_{2.5}$ emissions into context by considering the fraction of PM expected to be from HDV as a function of emission factor ratios. Ultimately, the validity of assumption that HDV contribute the bulk of aerosol is dependent on the ratio between HDV and LDV emission factors. In Fig. R1, we show the percentage of vehicle PM we expect to be from HDV on a highway as a function of HDV% for three scenarios: EFratio = 1, EFratio = 8, EFratio = 10, EFratio = 100, where the EFratio is defined as $EF_{PM(HDV)}$ / $EF_{PM(LDV)}$. Note that even when EFratio = 1, because emissions factors are in g PM / kg fuel, the percent of PM from HDV rises faster than HDV % because HDV burn 3-4x fuel per distance traveled. In 2009, we expect real emissions to fall between the EFratio = 10 and EFratio = 100, while in 2020, we expect real emissions to be closer to the EFratio = 8 line.

Finally, we feel that the evidence we present in Figure 5: a linear correlation between PM:CO enhancement ratio and HDV fraction with a very small intercept, and an estimated change in emission factors resulting from the introduction of a constant LDV emission factor, show that our assumptions are reasonable.

Concern 2: Goodness of fit for PM:CO enhancement ratios

Referee 1 is concerned about the statistics of the fitting we use in finding the ratio of PM enhancement to CO enhancement. Particularly, they are concerned about low R^2 values for our fits. We agree, as reviewer 1 suggests, that on an hourly basis, there is substantial uncertainty in calculating enhancements of PM and CO. We also concede that using a 68% CI to estimate uncertainty in the slopes we find to estimate error in emission factors may provide an overly optimistic picture of certainty in our estimates of emission factors. To address this, we have replaced the 68% CI with a 95% CI in slope to estimate the uncertainty in the fits used to estimate emission factors and carry this larger (95% CI) uncertainty for each slope through the process used to estimate emission factors.

However, we disagree with Referee 1's view that the noise in our data makes the fits we find essentially meaningless and totally uncertain. The information contained in this noisy data can be seen more clearly in considering median PM enhancement for bins of CO enhancement. When we fit binned medians, instead of all points, R^2 values increase substantially (Table R1), highlighting the information contained in this noisy data. Furthermore, when these binned medians are used to estimate emission factors, all emission factors agree to within error of emission factors estimated using all points (Table R2). Note that these values are slightly different than the table reported in the previous round of responses, as the median fit values from the previous round used all data before wind filtering and we now use data that has had a wind filter applied for both fit types. In the manuscript, we retain the “all points” fit as it was requested by reviewer 2, in the previous iteration of reviews.

We also note that R^2 values are worse when considering lower slopes. However, we find several cases in which R^2 values are relatively low, even for the binned median fits, but points fit the line well. See example in Figure R2. For fits to low slopes, and consequently small estimated emission factors, our estimated uncertainty in our factors is often large relative to the size of the estimated emission factor, and we believe the confidence interval of the slopes and consequent uncertainty estimates in emission factors to be reasonable.

Response to Referee 2:

We now describe the shaded region in Figure 5. In both Figure 1 and Figure 5, shaded region represents an error estimate for the blue line showing emissions factors from this study calculated by using error in quadriture.

References*:

* Carbotech, 1999 and Westerlund, 2001 were cited in a table EPA, 2020’s *Brake and Tire Wear Emissions from Onroad Emissions for MOVES3,* but we were unable to find the original reference.

Abu-Allaban, M., Gillies, J.A.,Gertler,A.W., Clayton ,R., Proffitt,D. (2002). Tailpipe, re-suspended road dust, and brake wear emission factors from on-road vehicles. AtmosphericEnvironment, 37(1),5283-5293.

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Habre, R., Girguis, M., Urman, R., Fruin, S., Lurmann, F., Shafer, M., Gorski, P., Franklin, M., McConnell, R., Avol, E. and Gilliland, F., 2021. Contribution of tailpipe and non-tailpipe traffic sources to quasi-ultrafine, fine and coarse particulate matter in southern California. *Journal of the Air & Waste Management Association*, *71*(2), pp.209-230.

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SENCO (Sustainable Environment Consultants Ltd.) (2000). Collation of information on particulate pollution from tyres, brakes, and road surfaces. 23 March, 1999, Colchester, Essex, UK

Rauterberg-Wulff A. (1999). Determination of emission factors for tyre wear particles up to 10μm by tunnel Measurements. Proceedings of 8th International Symposium ‘Transport and Air Pollution’.

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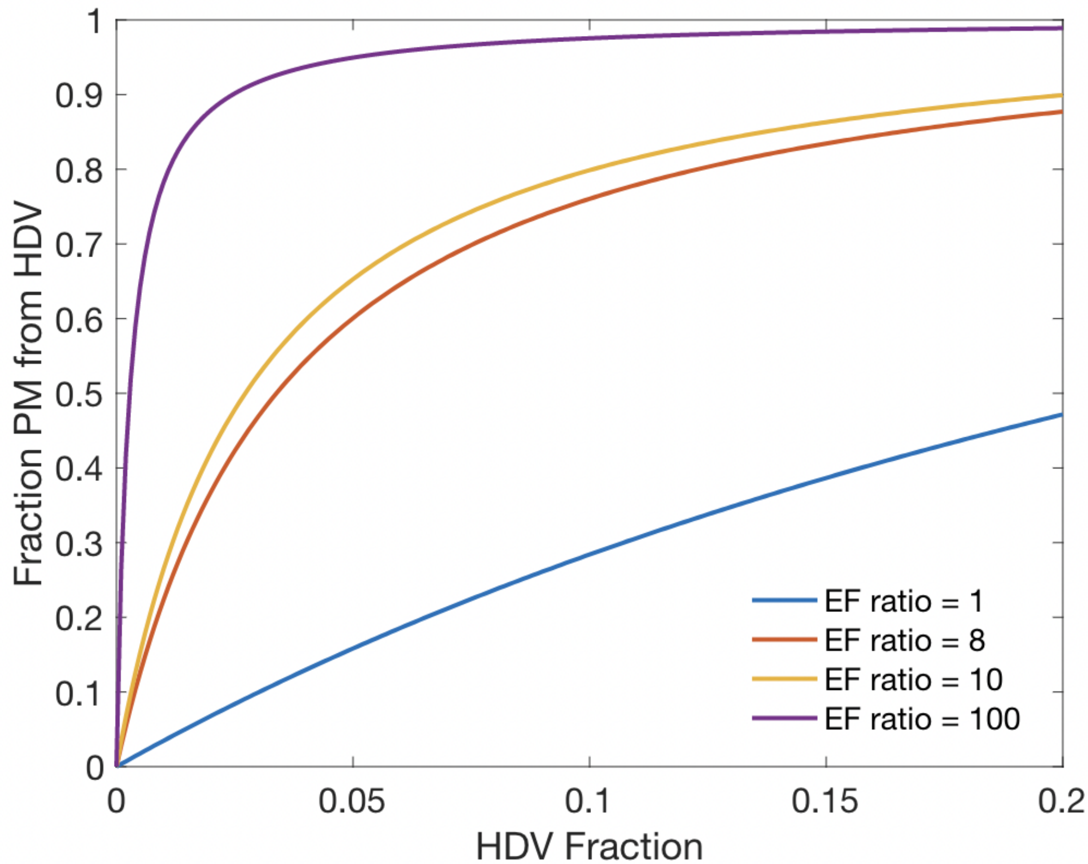

Figure R1: Expected fraction of PM from HDV (compared to total PM), for four different EF ratios, assuming fuel efficiencies 25 mpg for LDV and 7 mpg for HDV.

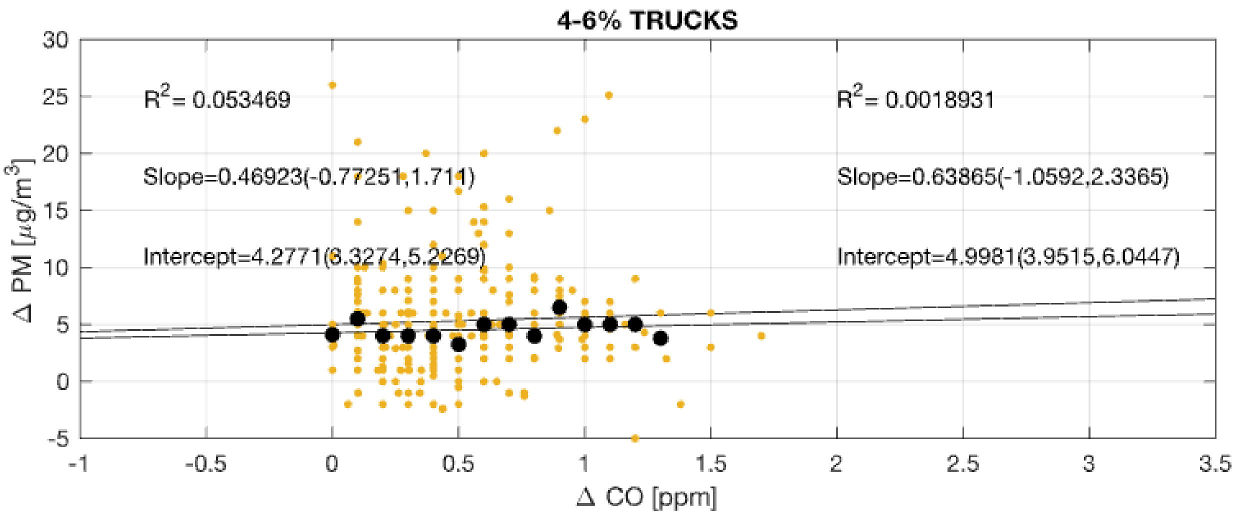

Figure R2: PM and CO enhancements for Redwood City in 2015-2027 for 4-6% trucks. Yellow points represent hourly data. Black dots represent binned medians. Fit values on the left represent fit to binned medians. Fit values on the right represent fit to all points. Numbers in parentheses represent 95th confidence intervals for all fit parameters.

	San Rafael		Redwood City		Berkeley Marina		Pleasanton	
2009 - 2011	*0.02*	*0.41*	*0.02* *0.03*	*0.02* *0.34*				
2012 - 2014	*0.10* *0.03*	*0.49* *0.77*	*0.06* *0.05* *0.06*	*0.29* *0.55* *0.19*				
2015 - 2017	*0.01* *0.003*	*0.09* *0.30*	*0.02* *0.002*	*0.22* *0.10*				
2018 - 2020	*0.0002* *0.03*	*0.51* *0.27*	*0.024* *0.002*	*0.68* *0.05*	*0.06*	*0.82*	*0.25* *0.31* *0.18*	*0.40* *0.86* *0.57*

Table R1: Here we report R^2 values for fits between PM enhancement and CO enhancement for each location and time period. Values on the left reflect R^2 for fits using all points, while those on right (yellow) reflect R^2 for fits using binned medians.

	San Rafael	Redwood City	Berkeley Marina	Pleasanton
2009 - 2011	1.10 +/- 0.86 **2.02 +/- 1.63**	0.31 +/- 0.17 **0.41 +/- 0.28**		
2012 - 2014	0.83 +/- 0.24 **0.72 +/- 0.29**	0.10 +/ 0.04 **0.15 +/- 0.06**		
2015 - 2017	0.41 +/- 0.21 **0.15 +/- 0.19**	0.05 +/- 0.05 **0.08 +/- 0.08**	0.49 +/- 1.80 —--	
2018 - 2020	0.11 +/- 0.11 **0.15 +/- 0.12**	0.05 +/- 0.06 **0.04 +/- 0.05**	0.15 +/- 0.05 **0.16 +/- 0.06**	0.35 +/- 0.08 **0.24 +/- 0.11**

Table R2: In this table, we calculate emission factor values calculated using either all points (top, not bold) or binned medians (**bottom, bold).** All values are in units g $PM_{2.5}$ / kg fuel.