



*Supplement of*

## **A method for using stationary networks to observe long-term trends of on-road emission factors of primary aerosol from heavy-duty vehicles**

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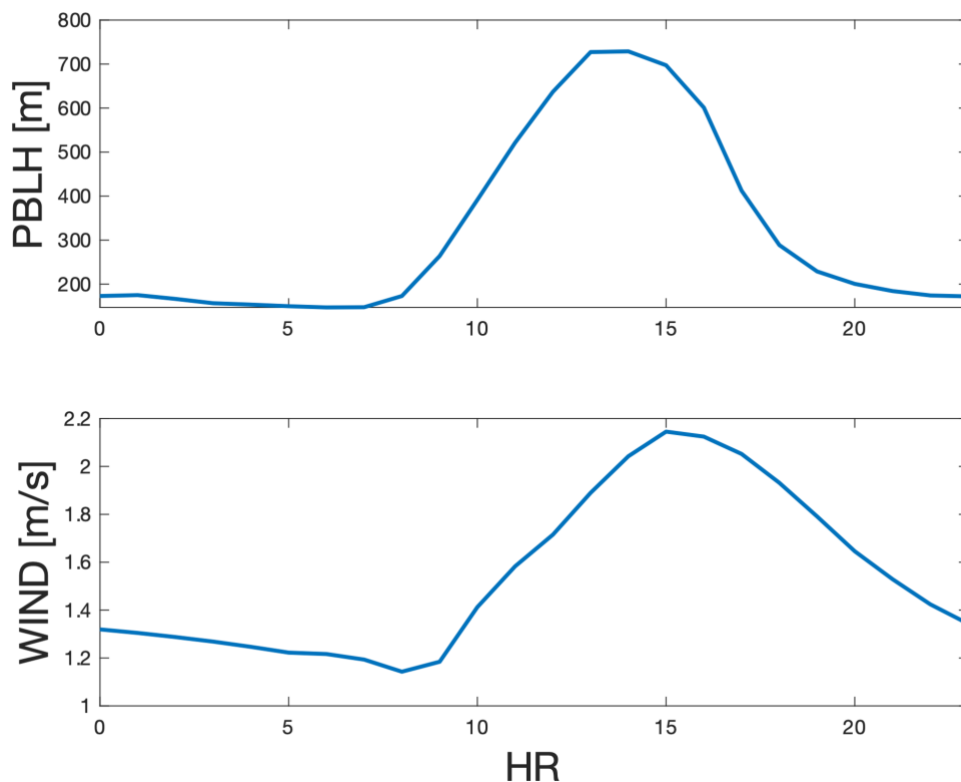
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## Introduction

In Sect. S1, we show typical diel of boundary layer height and windspeed in the area studied. Sect. S2 provides additional details on the Caltrans Performance Measurement System (PeMS), including a map of sites, IDs of sites used for data in this study, and typical flow HDV flow rates and HDV percent at sites studied. Section 3 provides a detailed derivation of Eq. 2 from the main text. In Sect. 4, we examine the sensitivity of our method to the time-window and percentile used in calculating baseline (regional)  $PM_{2.5}$  and CO concentrations. Section S5 shows linear fits between  $PM_{2.5}$  and CO enhancements for all sites and time periods. Section S6 details how we calculate uncertainty in final emission factor values. Section S7 shows an example of modeled  $PM_{2.5}$  enhancement. In Sect. S8, we use  $PM_{2.5}$ :CO enhancement ratios to calculate an emission factor at the Laney College site and compare computed factors to what would be expected from HDV alone as well as a contribution from the parking lot in which the site is located.

### Section S1: Meteorology Used in Modeled PM Enhancement

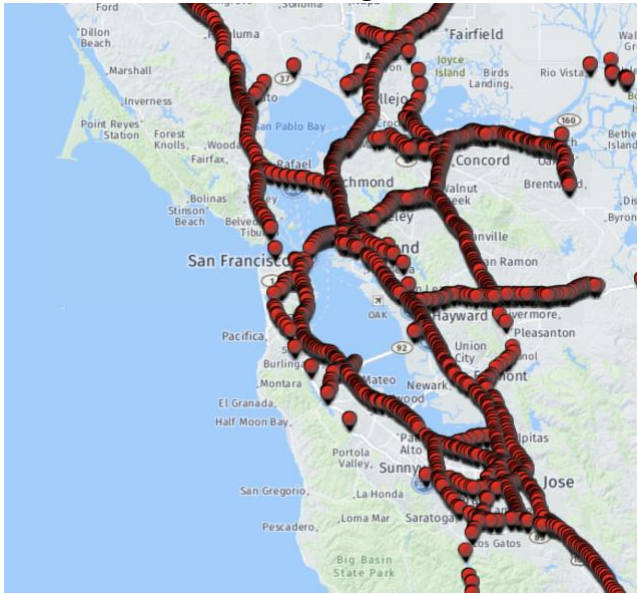
In the main text, we use wind speed and direction to filter  $\text{PM}_{2.5}$  and CO enhancements. We also use both wind speed and boundary layer height to make estimates of near roadway enhancement, using the continuity equation and gaussian plume dispersion. Here, we show diel cycles of the meteorology used in these calculations. Meteorological variables were taken from ECMWF ERA5.



**Figure S1:** Mean diel cycle for total boundary layer height (top) and wind speed (bottom) in Bay Area during winter and spring. Data averaged across 2009-2018.

## Section S2: Transportation Data

The Caltrans Performance Measurement System consists of a network of in-road sensors (magnetic loop) that detect car and truck flow rate across the state of California. PeMS derives truck portion at a given site using vehicle length estimates (Kwon, 2003). Comparisons of this method with weigh-in-motion technology finds error in this method to be ~5.7%. (Kwon, 2003) Although network density varies across the state of California, coverage in the San Francisco Bay Area is quite dense, with over 1800 measurement sites along major highways (Figure S1). Total vehicle flow rate and truck percentage were retrieved from (<http://pems.dot.ca.gov>). For each near-highway BAAQMD site, traffic data was taken from the two closest (primary) PeMS sites (one in either direction). In cases when PeMS data from the closest sites were not available, data was (if possible) filled in with median values for hour of week for the given site and year (excluding 2020), or retrieved from pairs of second closest (secondary) or third closest (tertiary) sites on the same highway. PeMS site codes in Table S1. Example flow rates are shown below in Figure S3.



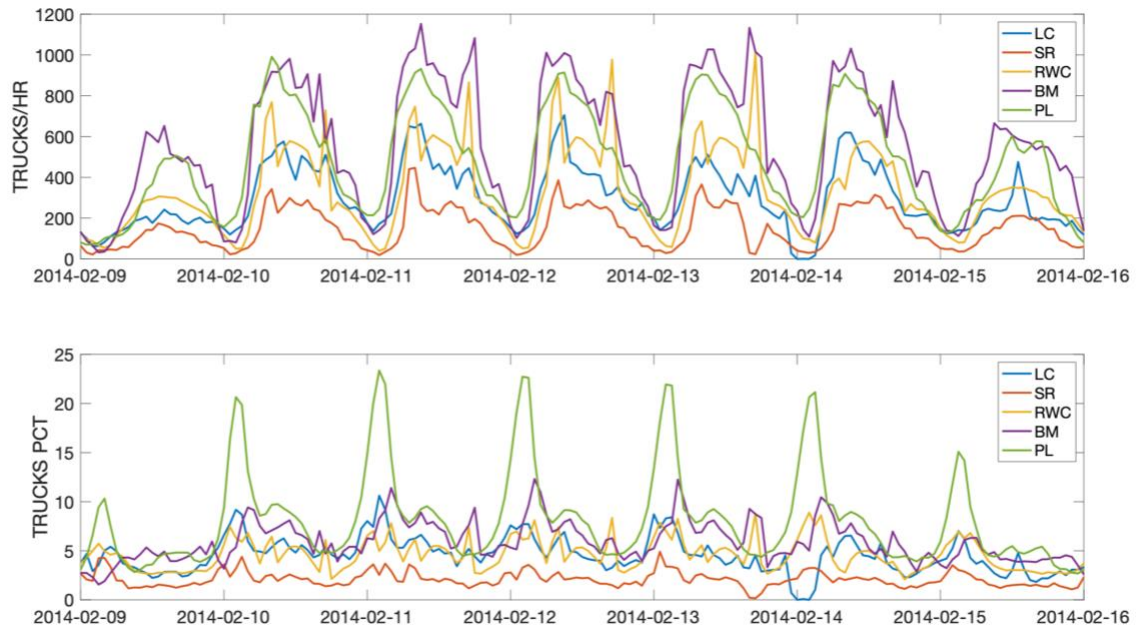
**Fig S2:** Map of Caltrans PeMS loop detector sites in the SF Bay Area from <http://pems.dot.ca.gov>. Copyright © 2022 State of California.

BAAQMD Site	PeMS – DIR 1 (primary)	PeMS – DIR 2 (primary)	PeMS – DIR 1 (secondary)	PeMS – DIR 2 (secondary)	PeMS – DIR 1 (tertiary)	PeMS – DIR 2 (tertiary)
Laney College (LC)	408138	400835	400609	400980	401710	400682
San Rafael (SR)	403317	403316	403314	403315	402412	402139
Redwood City (RWC)	404572	405673	401875	401874	401873	405679
Berkeley (BM)	400176	400728	400009	400432		
Pleasanton (PL)	402016	401006	400892	402018	402444	407964

**Table S1:** PeMS stations used in this study to capture truck flow near BAAQMD sites.

**Example weekly truck flow and truck percent at sites of interest:**

Hourly flow rates and truck percent are found by combining data from paired traffic sensors in each direction of flow. Peak weekday flow rates vary substantially from site to site from ~300 to ~1000 trucks / hr.



**Figure S3:** Hourly truck flow and truck % for PeMS sites located closes to the near-highway BAAQMD sites below.

### Section S3: Deriving Equation 2.

To derive Equation 2, we start with the definition of the HDV PM emissions factor in which the unit of activity is fuel burned:

$$EF_{PM,HDV} = \frac{g PM_{HDV}}{kg fuel_{HDV}}.$$

We multiply this expression by  $\frac{g CO_{fleet}}{g CO_{fleet}}$  and  $\frac{kg fuel_{fleet}}{kg fuel_{fleet}}$ , getting:

$$EF_{PM,HDV} = \frac{g PM_{HDV}}{kg fuel_{HDV}} \frac{g CO_{fleet}}{g CO_{fleet}} \frac{kg fuel_{fleet}}{kg fuel_{fleet}}.$$

Rearranging, we find:

$$EF_{PM,HDV} = \frac{g PM_{HDV}}{g CO_{fleet}} \frac{g CO_{fleet}}{kg fuel_{fleet}} \frac{kg fuel_{fleet}}{kg fuel_{HDV}}, \text{ so}$$

$$EF_{PM,HDV} = \frac{g PM_{HDV}}{g CO_{fleet}} \frac{g CO_{fleet}}{kg fuel_{HDV}}.$$

Because we measure concentrations of  $PM_{2.5}$  ( $\mu g m^{-3}$ ) and CO (ppm) rather than g PM emitted and g CO emitted, we convert using the ideal gas law.

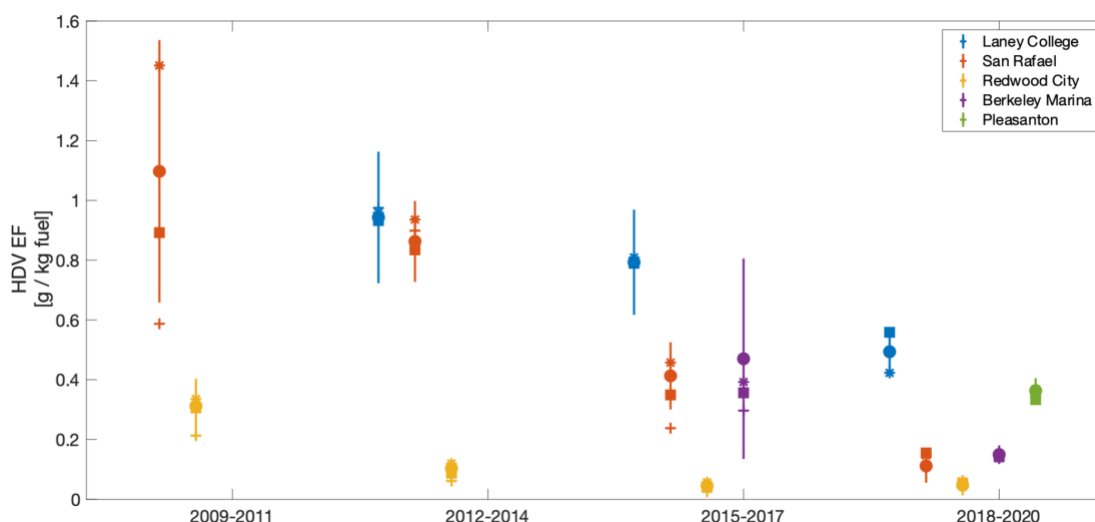
$$EF_{PM,HDV} = \gamma \frac{PM_{HDV}}{CO_{fleet}} \frac{g CO_{fleet}}{kg fuel_{HDV}}.$$

We calculate  $\gamma$  is using the ideal gas law, assuming STP.

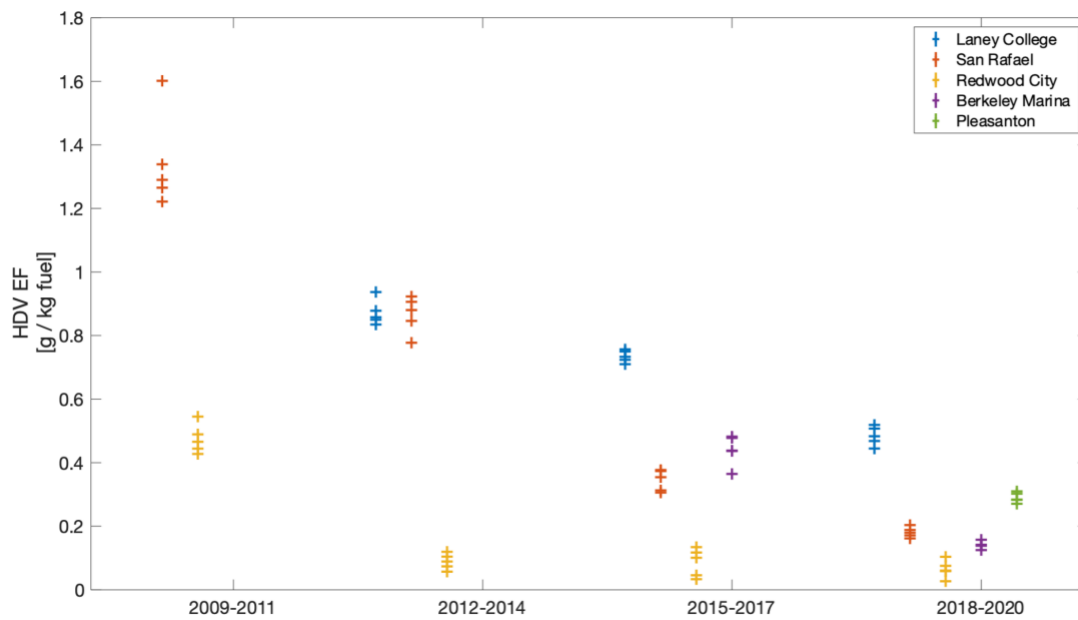
## Section S4: Sensitivity of Results to Regional Signal Method

We define regional signal of  $\text{PM}_{2.5}$  as including  $\text{PM}_{2.5}$  transported to the Bay Area from elsewhere,  $\text{PM}_{2.5}$  emitted from area point, and line sources far enough away from a site to have mixed through the area, and  $\text{PM}_{2.5}$  formed in the atmosphere through secondary chemical processes. We make the assumption that by taking the 10<sup>th</sup> percentile of the signal from all sites, that we are able to approximate this regional signal, as in Shusterman (2018). By subtracting the regional signal from total signal at a given site, we are able to isolate enhancements that result from localized emissions. We furthermore make the assumption that within the nearfield of a highway during morning rush hour, that both  $\text{PM}_{2.5}$  and CO enhancements are dominated by highway emissions.  $\text{PM}_{2.5}$  emissions not from the highway should not correlate well with enhancements in CO and are eliminated from our analysis by taking the median value of  $\text{PM}_{2.5}$  enhancement for each CO bin.

We choose to take a the 10<sup>th</sup> percentile of a five hour window, based on the size of the region we are trying represent, but we recognize that depending on meteorology, different time windows may be more appropriate. In Figure S3, we explore the sensitivity of emissions factors to the time window used to derive regional signal. In Figure S4, we explore the sensitivity of the emissions factors to the percentile used to derive regional signal. While we observe some dependence of HDV emissions factors on time window and percentile, we note that with the exception of San Rafael in 2009-2011, (1) temporal trends for a given site are unchanged, and (2) the spatial pattern of differences in emissions factors for a given time period are unchanged.



**Figure S4:** HDV emissions factors derived at each site during each time period, as in Figure 4 of the main text. Colors denote BAAQMD site: yellow denotes San Rafael, purple denotes Redwood City, blue denotes Laney College, and red denotes Berkeley. Each symbol represents a different time window used to derive regional signal: plus denotes one hour, square denotes three hours, circle denotes five hours, and the asterisk denotes seven hours. Error bars denote error calculated for 5 hour window.

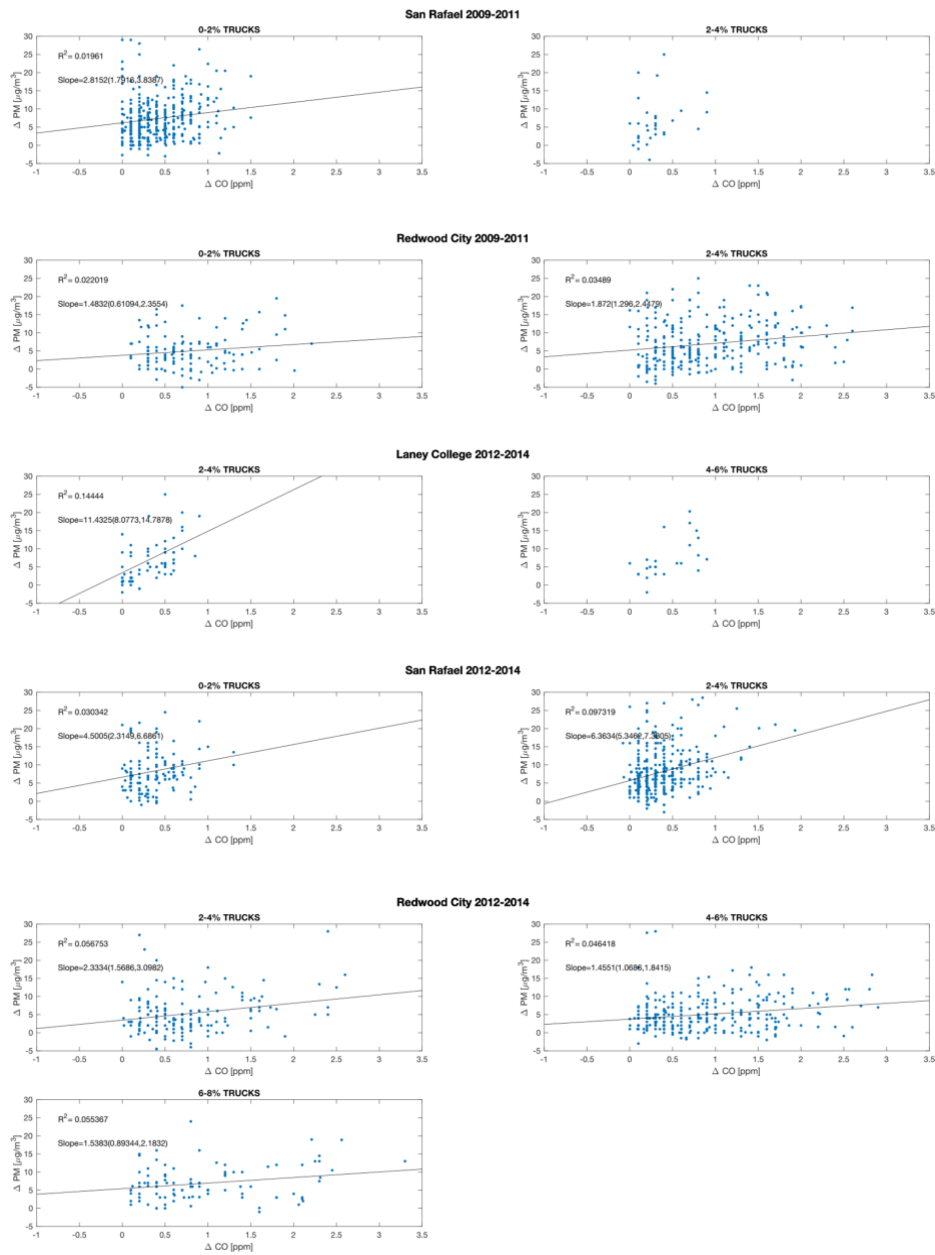


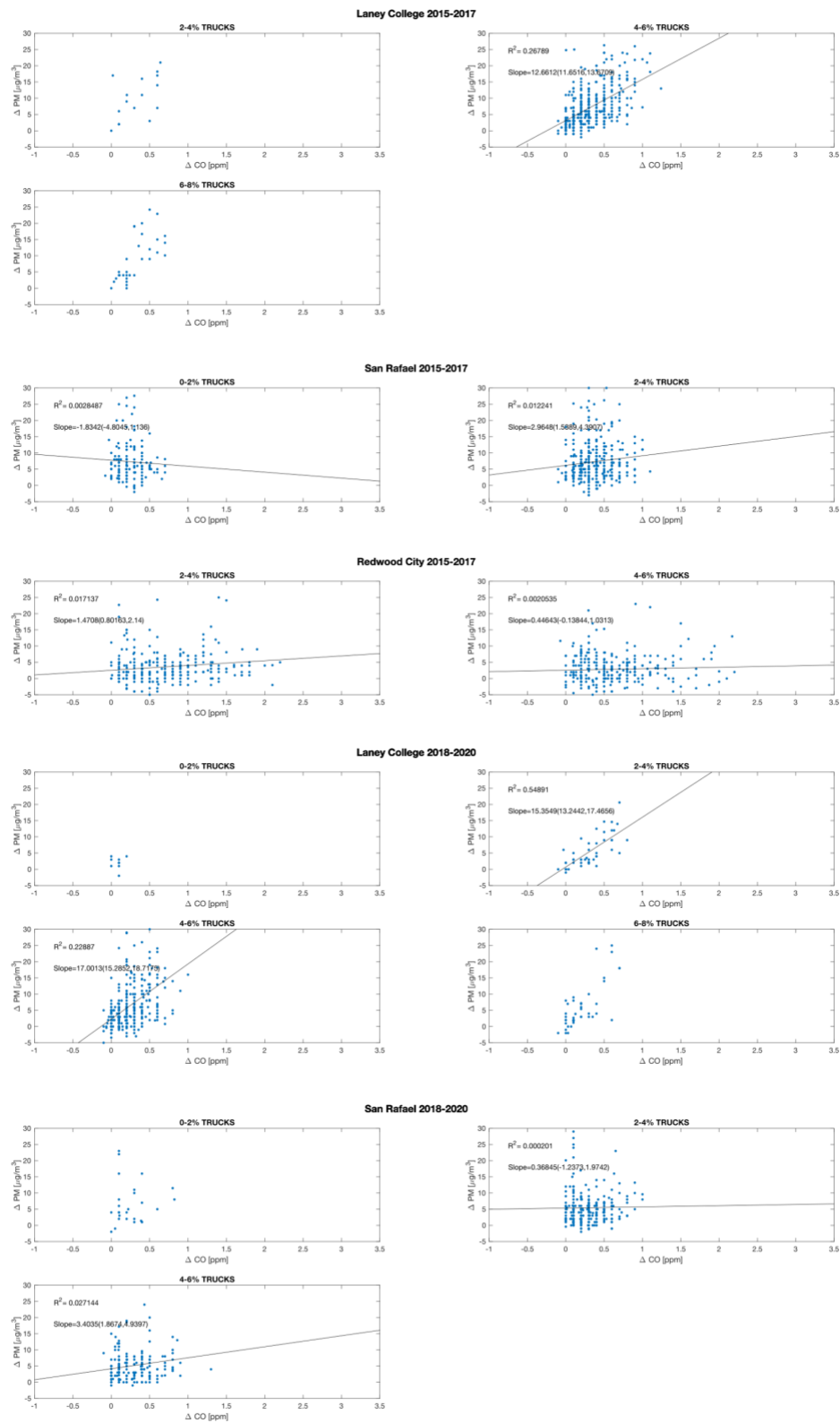
**Figure S5:** HDV emission factors derived at each site during each time period, as in Figure 4 of the main text. For each site and time period, emission factors were calculated using a five hour time window, but using different percentiles (5<sup>th</sup>, 10<sup>th</sup>, 15<sup>th</sup>, 20<sup>th</sup>, 25<sup>th</sup>) to estimate background values.

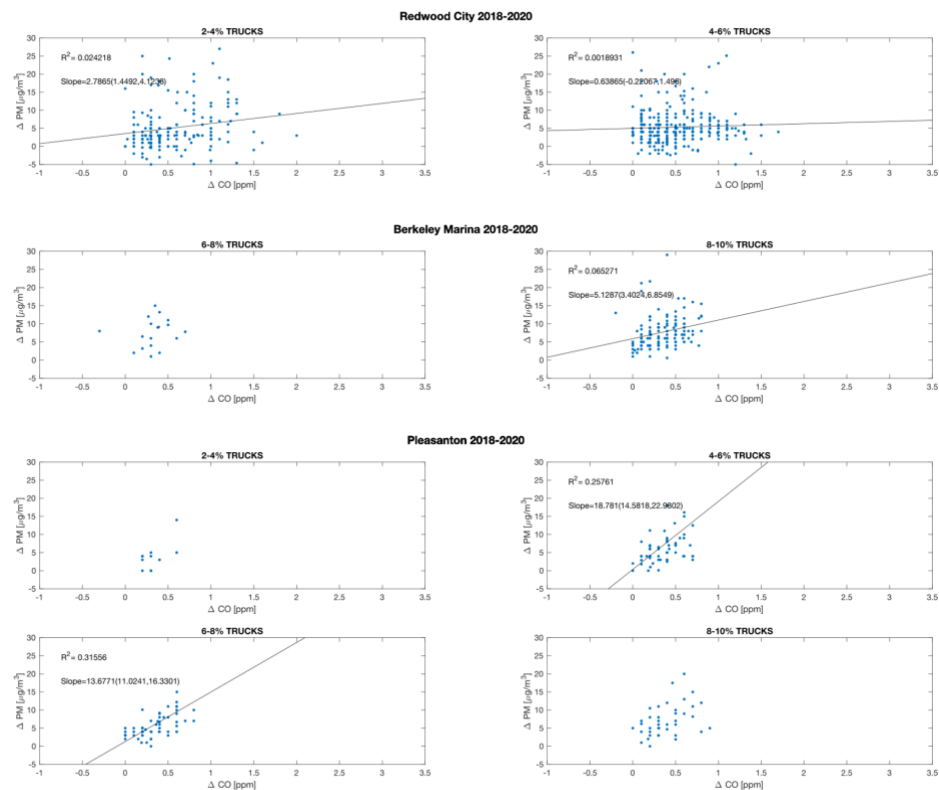


## Section S5: Determining Emissions Factors

As described in the main body of the text, we use enhancement in local CO over background as a tracer for PM<sub>2.5</sub> emitted by HDV on the highway. Although most of the CO comes from LDV, when averaging over the course of an hour, PM<sub>2.5</sub> emissions from HDV and CO emissions from LDV and HDV can be thought of as originating from the same location and can be thought to have the same, meteorologically dependent dilution from the roadway. Using our knowledge of truck percentage and total flow rate from PeMS and assuming a fleet-wide emissions factor for CO from HDV and LDV, we use enhancement ratios of PM<sub>2.5</sub> to CO to find HDV emissions factors for PM<sub>2.5</sub>, as described in the main text. Here we show the linear fits to enhancements for each site and time period.







**Figure S6:** Linear fits between PM enhancement and CO enhancement.

## Section S6: Uncertainties Related to Terms in Equation 2 and Equation 3

Equation 2 in the main text is given as:

$$EF_{PM,HDV} = \gamma \frac{PM_{HDV}}{CO_{fleet}} \frac{g CO_{fleet}}{kg fuel_{HDV}}.$$

We consider the ratios  $\frac{PM_{HDV}}{CO_{fleet}}$ , and  $\frac{g CO_{fleet}}{kg fuel_{HDV}}$  to be individual terms. Using error in quadrature, we find the uncertainty in  $EF_{PM,HDV}$  to be:

$$\delta EF_{PM,HDV} = \left\{ \left( \gamma \frac{g CO_{fleet}}{kg fuel_{HDV}} \delta \frac{PM_{HDV}}{CO_{fleet}} \right)^2 + \left( \gamma \frac{PM_{HDV}}{CO_{fleet}} \delta \frac{g CO_{fleet}}{kg fuel_{HDV}} \right)^2 \right\}^{1/2}.$$

We estimate  $\delta \frac{PM_{HDV}}{CO_{fleet}}$  using uncertainty in the fitting of local PM enhancement to local CO enhancement. Specifically, we estimate  $\delta \frac{PM_{HDV}}{CO_{fleet}}$  to be equal to half the difference between the 95% CI estimates of the slope of these fits. Because  $\frac{g CO_{fleet}}{kg fuel_{HDV}}$  is estimated using emissions rates and emissions factors from the EMFACA2017 model, we estimate their uncertainty by considering how these quantities vary within the model according to on road factors. Because the sites used in this study all correspond to roadways without significant grade, the largest contributor to uncertainty in these values is related how they change with vehicle speed, and so we find  $\delta \frac{g CO_{fleet}}{kg fuel_{HDV}}$  by finding speed-based variances in  $EF_{CO}$  and  $E$ .

Equation 2 in the main text is given as:

$$\frac{g CO_{fleet}}{kg fuel_{HDV}} = \frac{EF_{CO(HDV)}tE_{HDV} + EF_{CO(LDV)}(1-t)E_{LDV}}{tE_{HDV}}.$$

Thus, by using error in quadrature, we find that the uncertainty of  $EF_{CO,fleet}$  can be calculated as follows:

$$\delta \frac{g CO_{fleet}}{kg fuel_{HDV}} = \left\{ \delta EF_{CO(HDV)}^2 + \left( \frac{(1-t)E_{LDV}}{tE_{HDV}} \delta EF_{CO(LDV)} \right)^2 + \left( \left[ \frac{EF_{CO(HDV)}E_{HDV} + EF_{CO(LDV)}E_{LDV}}{tE_{HDV}} - \frac{EF_{CO(HDV)}tE_{HDV} + EF_{CO(LDV)}(1-t)E_{LDV}}{t^2E_{HDV}} \right] \delta t \right)^2 + \left( \left[ \frac{EF_{CO(HDV)}}{E_{HDV}} - \frac{EF_{CO(HDV)}tE_{HDV} + EF_{CO(LDV)}(1-t)E_{LDV}}{tE_{HDV}^2} \right] \delta E_{HDV} \right)^2 + \left( \frac{EF_{CO(LDV)}(1-t)}{tE_{HDV}} \delta E_{LDV} \right)^2 \right\}^{1/2}.$$

Running EMFAC2017 for the Bay Area Air Quality Management District region, we find specific CO emissions factors (g CO / kg fuel) and emissions rates (g CO<sub>2</sub> / mile) for each 5 mph speed bin for HDV and LDV separately. To do this, we classify EMFAC vehicle types as HDV or LDV based on

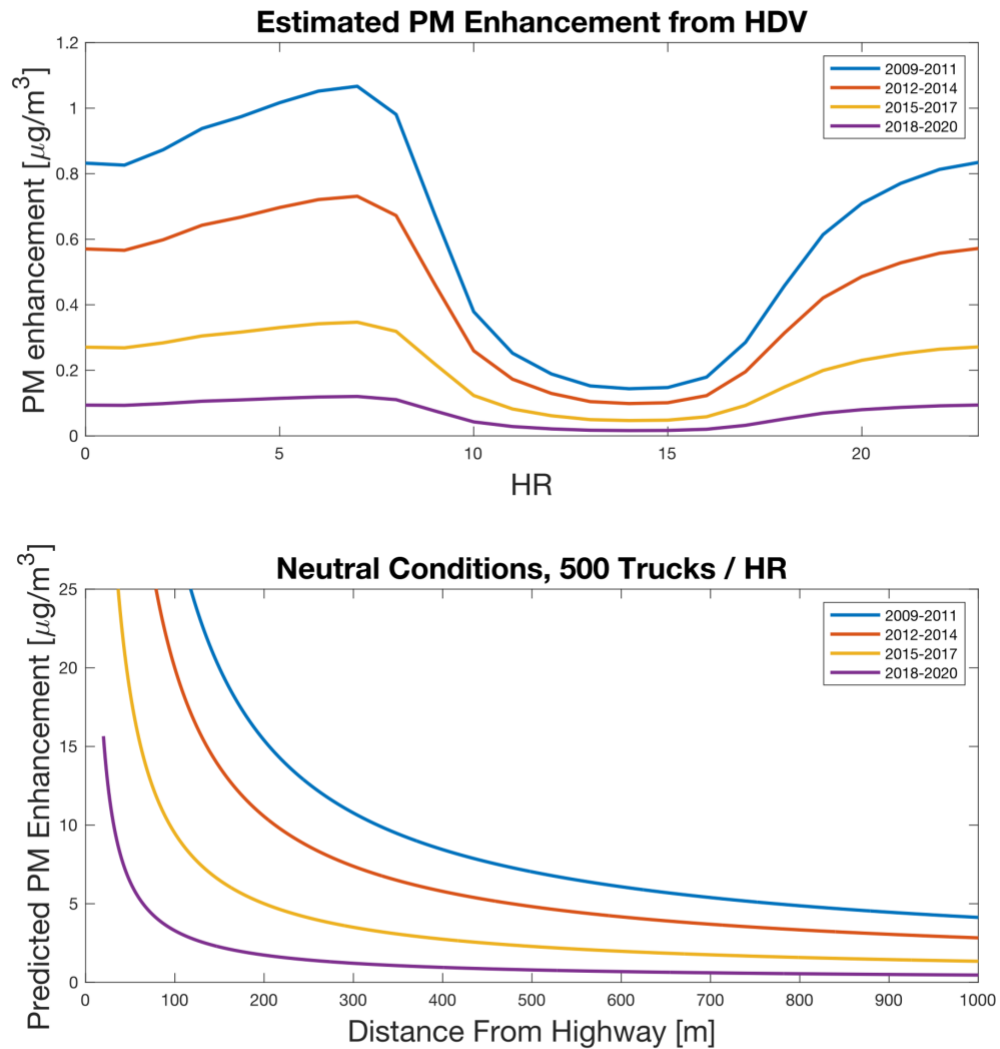
length, as listed in Fitzmaurice (2022). We then use PeMS speed data to estimate speed-dependent HDV and LDV emissions rates for each hour of data used find HDV emission factors. These values are found separately for each year as emission rates and factors change yearly in EMFAC2017 output. We estimate uncertainty in emission rates (g CO<sub>2</sub> / km) and CO emission factors (g CO / kg fuel) over a time period (e.g. 6-8am, weekdays, 2009-2011) by finding the standard deviation of EMFAC-PeMS-derived, speed-dependent emission rates and CO emission factors.

To estimate an uncertainty value for  $t$ , we refer to Kwon et al. (2003), who report that HDV volumes are accurate to within 5.7%. We can apply this to PeMS data to estimate  $\delta t$  to be:

$$\delta t = .057t.$$

## Section S7: Modeled PM<sub>2.5</sub> Enhancement

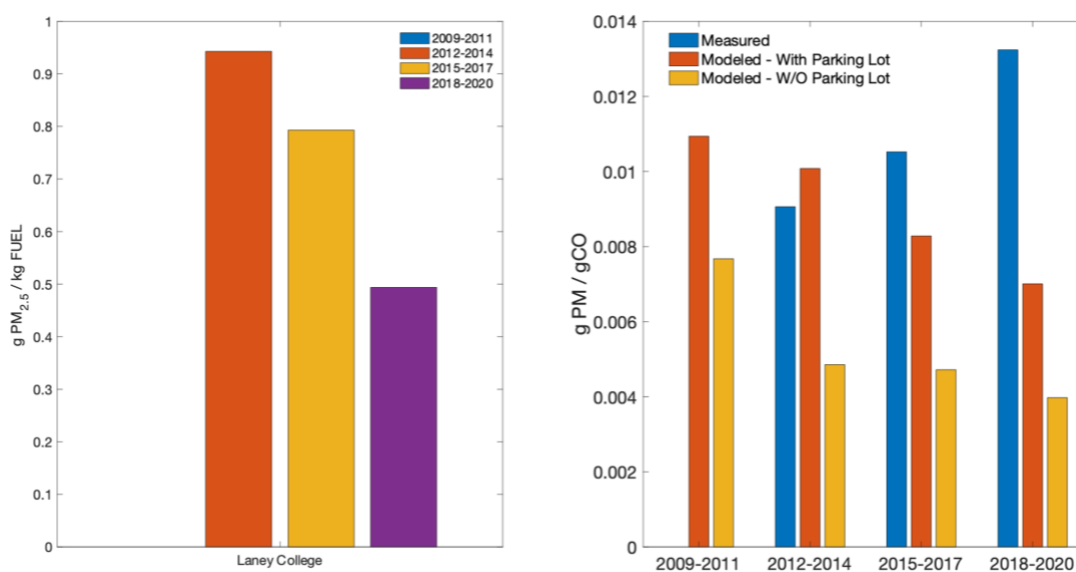
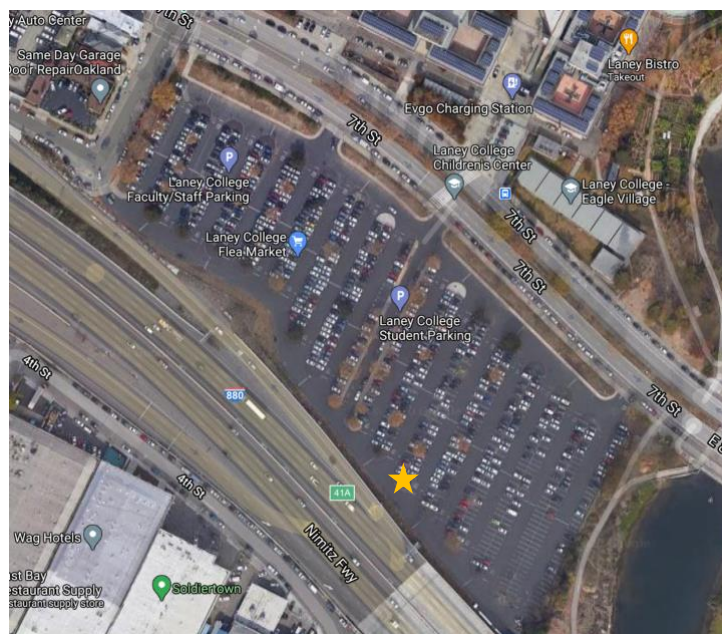
As described in the text, we model PM enhancement from HDV across the region and as a function of distance from the highway.



**Figure S7:** Modeled region-wide PM enhancement across Bay Area (top) and as a function of distance from a highway (bottom) during neutral conditions.

**Section S8:** Laney College is a near-highway BAAQMD site located in a large parking lot. We calculate much larger  $EF_{PM(HDV)}$  than for other sites (Fig. S8, bottom-left). We believe that this is due, at least in part, to emissions from the parking lot. EMFAC predicts PM:CO emissions ratio from LDV is expected to be substantially (~40x) higher at very low (5 mph) speeds, meaning that LDV in the parking lot will contribute substantially to PM enhancement and little to CO enhancement compared to highway LDV. Furthermore, the EMFAC model predicts that LDV moving at 5 mph have PM emission factors (g  $PM_{2.5}$  / kg fuel) of 72-96% and emission rates (g  $CO_2$  / km) of 100-109% of emission factors and emission rates of HDV driving at 50 mph. (Ranges in percentage indicate differences in time periods studied.) This means that an LDV driving through the parking lot would emit a similar amount of g  $PM_{2.5}$  as an HDV driving at highway speeds.

To demonstrate this idea, we use EMFAC2017 emissions factors for PM and CO for both LDV and HDV, as well as typical 7 am LDV and HDV flow at that site to predict a PM:CO ratio due to highway traffic alone as well as the ratio that is expected from highway traffic plus 650 cars per hour driving into the parking lot at 5mph (Fig. S8, bottom-right). EMFAC2017 predicts  $EF_{PM(LDV)}$  to be much higher at very low speeds, resulting in a substantially enhanced PM:CO ratio, that do not match, but are closer to the values measured at this site. This case highlights the need to screen near-highway sites for interfering emissions and the need to assess the role of slow moving LDV for their contribution to primary PM emissions.



**Figure S8:** (top) Aerial photo of parking lot in which Laney College AQ sensors located. Image retrieved from google maps (© Google Maps 2021). Yellow star indicates location of BAAQMD sensors. (bottom left)  $EF_{PM(HDV)}$  calculated by applying the procedure described in the text at Laney College. (bottom right) PM:CO ratios at Laney College site that are measured, modeled to include highway emissions only, or modeled to include both highway and parking lot emissions.



**References:**

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