Assessing vehicle fuel efficiency using a dense network of CO₂ observations

Helen L. Fitzmaurice¹, Alexander J. Turner², Jinsol Kim¹, Katherine Chan³, Erin <u>R.</u> Delaria⁴, Catherine Newman⁴, Paul 5 Wooldridge⁴, Ronald C. Cohen^{1,4}

J. Department of Earth and Planetary Science, University of California Berkeley, Berkeley, CA, 94720, United States,

2. Department of Atmospheric Sciences, University of Washington, Seattle, WA, 98195, United States,

3. Sacramento Metro Air Quality Management District, Sacramento, CA, 95814, United States,

4. Department of Chemistry, University of California Berkeley, Berkeley, CA, 94720, United States

10 Correspondence to: Ronald C. Cohen (rccohen@berkeley.edu)

Abstract. Transportation represents the largest sector of anthropogenic CO₂ emissions in urban areas in the United States. Timely reductions in urban transportation emissions are critical to reaching climate goals set by international treaties, national policies, and local governments. Transportation emissions also remain one of the largest contributors to both poor air quality (AQ) and to inequities in AQ exposure. As municipal and regional governments create policy targeted at reducing

- 15 transportation emissions, the ability to evaluate the efficacy of such emission reduction strategies at the spatial and temporal scales of neighborhoods is increasingly important. However, the current state of the art in emissions monitoring does not provide the temporal, sectoral, or spatial resolution necessary to track changes in emissions and provide feedback on the efficacy of such policies at a neighborhood scale. The BErkeley Air Quality and CO₂ Network (BEACO₂N) has previously been shown to provide constraints on emissions from the vehicle sector in aggregate over a ~1300 km² multi-city spatial
- 20 domain. Here, we focus on a 5 km, high volume, stretch of highway in the SF Bay area. We show that inversion of the BEACO₂N measurements can be used to understand two factors that affect fuel efficiency: vehicle speed and fleet composition. The CO₂ emission rate of the average vehicle (g/vkm) are shown to vary by as much as 27% at different times of a typical weekday because of changes in <u>these two factors</u>. The BEACO₂N-derived emissions estimates are consistent to within ~3% of estimates derived from publicly available measures of vehicle type, number, and speed, providing direct 25 observational support for the accuracy of the Emissions FACtor model (EMFAC) of vehicle fuel efficiency.

1 Introduction

Urban emissions currently account for ~75 % of all anthropogenic CO₂ emissions (IPCC, 2014). By 2050, roughly two-thirds of the earth's projected population of 9.3 billion is expected to reside within urban areas (IPCC, 2014), meaning that effective greenhouse gas emissions reductions strategies must focus on urban emissions reductions.

Style Definition		
Style Definition [19] Style Definition [17] Style Definition [16] Style Definition [15] Style Definition [14] Style Definition [13] Style Definition [11] Style Definition [11] Style Definition [11] Style Definition [10] Style Definition [10] Style Definition [10] Style Definition [11] Style Definition [10] Style Definition [11] Style Definition [12] Style Definition [12] Style Definition [21] Style Definition [22] Formatted [22] Formatted [22] Formatted [23] Formatted [24] Deleted: [25] Deleted: [26] Deleted: [27] Deleted: [28] Formatted [28]	Style Definition	([21])
Style Definition (Style Definition	([20]
Style Definition (Style Definition	([19]
Style Definition (Style Definition	([18]
Style Definition	Style Definition	([17]
Style Definition (Style Definition	([16]
Style Definition ([13] Style Definition ([12] Style Definition ([10] Style Definition ([17] Style Definition ([21] Style Definition ([21] Style Definition ([22] Formatted ([22] Formatted ([22] Formatted ([22] Deleted: ([23	Style Definition	
Style Definition	Style Definition	[14]
Style Definition [11] Style Definition [10] Style Definition [10] Style Definition [17] Style Definition [16] Style Definition [17] Style Definition [17] Style Definition [13] Style Definition [21] Style Definition [22] Formatted [23] Formatted [24] Deleted: 1. University of California, Berkeley, Formatted [25] Deleted: University of Washington, Formatted [26] Deleted: [27] Deleted: [27] Deleted: [28] Formatted [28] Formatted [28] Formatted [28] Formatted [21] Deleted: (recohen@berkeley.edu) [28] <th>Style Definition</th> <th>([13]</th>	Style Definition	([13]
Style Definition (Style Definition	[12]
Style Definition	Style Definition	[11]
Style Definition	Style Definition	[10]
Style Definition	Style Definition	[9]
Style Definition ([1] Style Definition ([5] Style Definition ([3] Style Definition ([2] Style Definition ([2] Style Definition ([2] Formatted ([2] Deleted: 1. University of California, Berkeley, Formatted ([26] Deleted: University of Washington, Formatted ([27] Deleted: ([27] Deleted: ([27] Deleted: ([28] Formatted ([28] Formatted ([28] Formatted ([31] Deleted: (recohen@berkeley.edu) ([31] Deleted: ([32] Deleted: ([32] Deleted: ([33] Formatted ([34]	Style Definition	[8]
Style Definition [Style Definition	[7]
Style Definition	Style Definition	[6]
Style Definition	Style Definition	([5]
Style Definition	Style Definition	[4]
Style Definition [2] Style Definition [1] Formatted [22] Formatted [23] Formatted [24] Deleted: 1. University of California, Berkeley, Formatted [25] Deleted: 1. University of California, Berkeley, Formatted [25] Deleted: University of Washington, Formatted [26] Deleted:	Style Definition	([3]
Style Definition [1] Formatted [22] Formatted [23] Formatted [24] Deleted: 1. University of California, Berkeley, Formatted [25] Deleted: 1. University of Washington, Formatted [25] Deleted: University of Washington, Formatted [26] Deleted:	Style Definition	[2]
Formatted ([23] Formatted ([24] Deleted: 1. University of California, Berkeley, Formatted ([25] Deleted: University of Washington, Formatted ([26] Deleted: Formatted ([27] Deleted: , Formatted ([27] Deleted: , Formatted ([29] Deleted: Department of Chemistry Formatted ([28] Formatted ([28] Formatted ([30] Deleted: (recohen@berkeley.edu) ([31] Deleted: . ([31] Deleted: . ([32] Deleted: vehicle speed and fleet composition. ([33] Formatted ([34]	Style Definition	
Formatted ([24] Deleted: 1. University of California, Berkeley, Formatted ([25] Deleted: University of Washington, Formatted ([26] Deleted: University of Washington, Formatted ([26] Deleted: ([27] Deleted: , Formatted ([27] Deleted: , ([29] Deleted: Department of Chemistry Formatted ([28] Formatted ([28] Formatted ([30] Deleted: (recohen@berkeley.edu) ([31] Deleted: . ([31] Deleted: . ([32] Deleted: vehicle speed and fleet composition. ([33] Formatted ([33] Formatted ([34]	Formatted	[22]
Deleted: 1. University of California, Berkeley, Formatted Construction Formatted Construction Formatted Construction Formatted Construction Formatted Construction Construction Formatted Construction Construction Formatted Construction Construction Formatted Construction	Formatted	([23]
Formatted ([25] Deleted: University of Washington, ([26] Formatted ([26] Deleted: ([27] Deleted: ([29] Deleted: Deleted: Formatted ([28] Formatted ([30] Deleted: (rcohen@berkeley.edu) Formatted ([31] Deleted: ([31] Deleted: ([32] Deleted: vehicle speed and fleet composition. Formatted ([33] Formatted ([34]	Formatted	[24]
Deleted: University of Washington, Formatted Construct Formatted Construct Formatted Construct Construct Construct Construct Formatted Construct Construct<	Deleted: 1. University of California, Berkeley,	
Formatted ([26] Deleted: ([27] Deleted: ([27] Deleted: ([27] Deleted: ([29] Deleted: ([29] Deleted: ([29] Deleted: ([28] Formatted ([28] Formatted ([30] Deleted: (recohen@berkeley.edu) Formatted ([31] Deleted: ([32] Deleted: vehicle speed and fleet composition. ([33] Formatted ([34]	Formatted	([25]
Deleted: ([20] Formatted ([27] Deleted: . Formatted ([29] Deleted: Department of Chemistry [28] Formatted ([28] Formatted ([30] Deleted: (recohen@berkeley.edu) [30] Formatted ([31] Deleted: [31] Deleted: [32] Deleted: vehicle speed and fleet composition. [33] Formatted ([33] Formatted ([34]	Deleted: University of Washington,	
Formatted ([27] Deleted: . [29] Formatted ([29] Deleted: Department of Chemistry [28] Formatted ([28] Formatted ([30] Deleted: (recohen@berkeley.edu) [30] Deleted: (recohen@berkeley.edu) [31] Deleted: . [32] Deleted: vehicle speed and fleet composition. [33] Formatted ([34] Formatted ([34]	Formatted	[26]
Deleted: ([2]) Deleted: ([29]) Deleted: Deleted: Formatted ([28]) Formatted ([30]) Deleted: (rcohen@berkeley.edu) Formatted ([31]) Deleted: ([32]) Deleted: ([32]) Deleted: vehicle speed and fleet composition. ([33]) Formatted ([34])	Deleted:	
Formatted ([29] Deleted: Department of Chemistry Formatted ([28] Formatted ([30] Deleted: (recohen@berkeley.edu) ([30] Formatted ([31] Deleted: . ([32] Deleted: vehicle speed and fleet composition. ([33] Formatted ([34]		([27]
Deleted: Department of Chemistry Formatted Formatted ([28] Formatted ([30] Deleted: (recohen@berkeley.edu) Formatted Formatted ([31] Deleted: . Formatted ([32] Deleted: vehicle speed and fleet composition. Formatted ([33] Formatted ([34]		
Formatted ([28] Formatted ([30] Deleted: (rccohen@berkeley.edu) ([31] Formatted ([31] Deleted: . ([32] Deleted: vehicle speed and fleet composition. ([33] Formatted ([33] Formatted ([34]		([29]
Formatted ([30] Deleted: (rccohen@berkeley.edu) ([31] Formatted ([31] Deleted: . ([32] Deleted: vehicle speed and fleet composition. ([33] Formatted ([33] Formatted ([34]	Deleted: Department of Chemistry	
Deleted: (recohen@berkeley.edu) Formatted Geleted: Formatted ([31] Deleted: Formatted ([32] Deleted: vehicle speed and fleet composition. Formatted Formatted ([33] Formatted ([34]		([28]
Formatted ([31] Deleted: . Formatted ([32] Deleted: vehicle speed and fleet composition.	>	([30]
Deleted:	>	
Formatted ([32] Deleted: vehicle speed and fleet composition. Formatted ([33] Formatted ([34]		([31]
Deleted: vehicle speed and fleet composition. Formatted Formatted ([33]	<u></u>	
Formatted ([33] Formatted ([34] [34] <th[34]< th=""> <th[34< th=""><th></th><th>([32]</th></th[34<></th[34]<>		([32]
Formatted ([34]		
Formatted ([35]		
	Formatted	([35]

The transportation sector is responsible for \sim 23% of global greenhouse gas emissions worldwide (IPCC, 2014) and \leftarrow represents the greatest sectoral percentage (\sim 25-66%) of emissions from within the boundaries of urban areas in the United States (Daw, 2020; Gurney et al., 2021). Although fuel efficiency of new internal combustion engine vehicles has increased by \sim 30% over the last 20 years and electric vehicles (EV) are becoming more prevalent (e.g.

- 50 https://arb.ca.gov/emfac/emissions-inventory), emissions reductions resulting from fuel efficiency gains in newer vehicles are negated by an increasing percentage of heavy-duty vehicles (HDV) (Moua, 2020), speed-related reductions in fuel efficiency resulting from increases in congestion, and an increase of total vehicle kilometers traveled (vkm). Over the past 20 years, even in locations with aggressive climate change policy, these factors have resulted in CO2 emissions from vehicles that have increased or stayed nearly constant. For example, California Air Resources Board estimates that in the state of
- 55 California, per capita vehicle emissions in 2015 were only 2% lower than in 2000 and per capita vehicle kilometers traveled (vkm) increased ~2.5% over that time period (California Air Resources Board, 2018). In addition to GHG emissions, the transportation sector is responsible for a significant share of PM_{2.5} and NO_x emissions, exacerbating PM_{2.5} and ozone exposure in low-income communities and communities of color already experiencing disproportionate health burdens associated with poor air quality (Tessum et al., 2021).
- 60 Municipal and regional governments have increasingly shown interest in tracking and reducing CO_2 emissions from all sectors, including transportation. For example, Boswell et al₄(2019) found that 64% of Californians live in a city with a climate action plan. For urban and regional governments to plan, monitor, and responsively adjust emissions reduction policies, an up-to-date understanding of the spatial and temporal variations in total emissions and in emissions by sector and subsector processes is key.
- 65 For transportation, reductions in vkm, congestion mitigation, and rules affecting fleet composition (e.g., limiting road access to HDV, incentivizing use of electric vehicles, or buy-backs of older vehicles) are three levers that can be employed to reduce CO₂ and AQ emissions from vehicles, thereby affecting the climate footprint, air quality (AQ), and environmental justice (EJ) in a region. However, the current state of the art in emissions monitoring and modelling do not provide the temporal, sectoral, or spatial resolution necessary to track changes in urban emissions and provide feedback on
- 70 the efficacy of each lever separately. Furthermore, current estimates of the magnitude and sectoral apportionment, of urban CO₂ emissions can vary widely. For example, Gurney et al. (2021) show how a consistent approach to total emissions from cities across the U.S. differs from locally constructed inventories in magnitude and sector by sector.

Spatial and temporal process-level maps of emissions are needed to improve the scientific basis for emission control strategies. The current state of the art involves finding aggregate emissions over large regions (counties, states) using

75 economic data and downscaling those totals using proxies such as road length, building type or population density. These j models meet the need for high spatial resolution (~500 m) and capture emissions from many detailed subsectors (Gately, et al., 2015; Gurney et al., 2012; McDonald, et al., 2014). Because fuel sales are well-characterized, these models are also likely to produce accurate region-wide CO₂ emissions totals from the transportation sector.

Formatted: Line spacing: 1.5 lines

Deleted: Kevin Robert

Fo	ormatted: Font: 10 pt
De	eleted: 2018
De	eleted: travelled
Fo	ormatted: Font: 10 pt
De	eleted: per capita
Fo	ormatted: Font: 10 pt
Fo	ormatted: Font: 10 pt

Formatted: Font: 10 pt Deleted: Boswell & Madilyn Jacobson,

Deleted: Furthermore, current estimates of the magnitude and sectoral apportionment, of urban CO_2 emissions can vary widely (3, 8). For example, Gurney et al. show how a consistent approach to total emissions from cities across the U.S. differs from locally constructed inventories in magnitude and sector by sector (Kevin Robert Gurney et al., 2021).

Formatted: Font: 10 pt
Deleted: traffic
Formatted: Font: 10 pt
Deleted: either vehicle flow rates or
Formatted: Font: 10 pt
Deleted: like
Formatted: Font: 10 pt
Deleted: and
Formatted: Font: 10 pt
Deleted: Conor K.
Deleted: , Hutyra, & Wing,
Deleted: Kevin R.
Deleted: , McBride, Martin, & Harley,
Deleted: .
Formatted: Font: 10 pt

Yet even the most detailed of these inventories do not presently describe the temporal variability in processes that affect emissions, such as the direct response of home heating or air conditioning to ambient temperature or, with one exception (Gately et al., 2017b), the variations in emissions per km when comparing free-flowing to stop-and-go traffic. These models often disagree with one another spatially (Gately et al., 2017a), have been subject to only limited testing, against observations of the atmosphere, and are not designed to be consistent with separately constructed AQ inventories that have been subject to much more extensive testing against observations.

- 105 Mobile monitoring campaigns and high-density measurement networks highlight the importance of characterizing and identifying the processes contributing to sharp neighborhood-scale AQ and GHG hotspots and point to the importance of traffic emissions on neighborhood scales. For example, Apte et al. (2017), showed that concentrations of NOx and Black Carbon (BC) can vary by as much as a factor of ~8 on the scale of 10s to 100s of meters. Caubel et al. (2019), showed BC concentrations to be ~2.5 times higher on trucking routes than on neighboring streets. Such gradients are not represented in
- 110 inventories based on downscaled economic data.

Observations of CO₂ and other greenhouse gases can play an important role in improving and maintaining the accuracy of emission models—especially during a time of rapid proposed changes. CO₂ measurements paired with Bayesian inverse models have been shown to provide a quantitative assessment of emissions (Lauvaux et al., 2016; Lauvaux et al., 2020; Turner, et al., 2020a). To date, most attempts at quantifying urban CO₂ emissions have focused on extracting a

115 temporally averaged (often a full year) total of the anthropogenic CO₂ across the full extent of city. A few studies have attempted to disaggregate emissions by sector or fuel type, or describe large shifts in aggregate emissions (Newman et al., 2016; Nathan et al., 2018; Lauvaux et al., 2020; Turner, et al., 2020a), but none characterize subsector processes of vehicle emissions.

High spatial density observations offer promise as a means to explore process-level emissions details. The BErkeley 120 Air Quality and CO₂ Network (BEACO₂N) is an observing network deployed in the San Francisco Bay Area and other cities with measurement spacing of ~2km (Fig. 1, left). In a prior analysis, Turner et al. <u>12020a</u>) showed that BEACO₂N measurements can detect variation in CO₂ emissions with time of day and day of week in addition to the dramatic changes in CO₂ emissions due to the COVID-related decrease in driving.

Here, we analyze hourly, spatially-allocated CO₂ emissions derived from the inversion of BEACO₂N observations (Turner, et al., <u>2020a</u>) to explore how well they constrain the CO₂ emissions from a 5km stretch of highway, <u>This stretch</u> chosen because of its location upwind of consistently active <u>BEACO₂N</u> sites and for completeness of traffic data, and <u>because emission rates</u> are highly affected by speed (vehicles use more fuel per km at very low and high speeds) and fleetcomposition (HDV emit more CO₂ per km than light duty vehicles (LDV)). The variation of the ratio of total fleet CO₂ emission per vehicle km traveled (g_CO₂/ykm) is used to explore variations in on-road fuel efficiency and the factors

130 responsible for that variation. We show that average fuel efficiency of the vehicle fleet on the road varies by as much as 27% over the course of a typical weekday.

Deleted: Conor K.

Deleted: , Hutyra, Peterson, & Sue Wing, 2017

Deleted: C. K.

Deleted: & Hutyra, 2017

Deleted: and for the most part, are not tested

Formatted: Font: 10 pt

Formatted: Font: 10 pt

Deleted: Mobile monitoring campaigns and high-density measurement networks highlight the importance of characterizing and identifying the processes contributing to sharp neighborhood scale AQ and GHG hotspots and point to the importance of traffic emissions on this scale. For example, Apte et al, showed that concentrations of NOx and BC can vary by as much as a factor of ~8 on the scale of 10s to 100s of meters (Ante et al. 2017). Caubel et al, showed BC concentrations to be ~ 2.5 times higher on trucking routes than on neighboring streets (Caubel, Cados, Preble, & Kirchstetter, 2019). Such gradients are not represented in inventories based on downscaled economic data. Observations of CO2 and other greenhouse gases can play an important role in improving and maintaining the accuracy of emission models-especially during a time of rapid proposed changes. CO2 measurements paired with Bayesian inverse models have been shown to provide a quantitative assessment of emissions (Lauvaux et al., 2020, 2016; Turner, Kim, et al., 2020). To date, most attempts at quantifying urban CO2 emissions have focused on extracting a temporally averaged (often a full year) total of the anthropogenic CO₂ across the full extent of city. A few studies have attempted to disaggregate emissions by sector or describe large shifts in aggregate emissions (Lauvaux et al., 2020; Turner, Kim, et al., 2020), but none characterize subsector processes.

Formatted: Font: 10 pt

Formatted: Line spacing: 1.5 lines

Deleted: Figure

Formatted: Font: 10 pt

Deleted: (Turner, Kim, et al., 2020)

(Deleted: , Kim,
~(Deleted: 2020
1	Deleted: where emissions
(Formatted: Font: 10 pt
(Formatted: Font: 10 pt
(Formatted: Font: 10 pt
-(Deleted: /
Y	Formatted: Font: 10 pt

2 Methods and Data

2.1 The Berkeley Air quality and CO2 Network

- 170 We use hourly CO₂ observations from the Berkeley Air quality and CO₂ Network (BEACO₂N) (Shusterman et al., 2016; <u>Kim et al., 2018; Delaria et al., 2021). The BEACO₂N network includes more than 70 locations in the SF Bay Area, spaced at ~2 km, and measures CO₂ with a network instrument error of 1.6 ppm or less (Delaria et al., 2021). All available data from January-June 2018-2020 are included in this analysis. During this time, more than 50 <u>distinct locations had nodes that were</u> active for a month or more (including 19 sites within 10 km of our highway stretch of interest). The number of nodes active</u>
- 175 at any given time ranged from 7-41, with a mean of 17. Figure 1 shows sites in operation at some point during analysis period and Fig. S1 shows a timeseries of the number of nodes available throughout the study period.

2.2 The BEACO₂N - STILT Inversion System

"To infer CO₂ emissions from within the BEACO₂N footprint, we use the Stochastic-Time Inverted Lagrangian Transport

- 180 (STILT) model, coupled with a Bayesian inversion as described in detail in Turner et al. (2020a). Briefly, we use meteorology from NOAA's HRRR product at 3 km resolution to calculate footprints from each hour at each site, weighted by a priori CO₂ emissions. The overall region of influence, the network footprint, as defined by a contour representing 40% of the CO₂ influence is shown in Fig. S2 (left). We construct a spatially gridded prior emissions inventory using point sources provided by the Bay Area Air Quality Management District (2011), home heating emissions as reported by
- 185 BAAQMD (2011) and distributed spatially according to population density, on-road emissions from the High-resolution Fuel Inventory for Vehicle Emissions (McDonald et al., 2014) varying by hour of week and scaled by year using fuel sales data, and a biogenic inventory derived using Solar Induced Fluorescence (SIF) Satellite data (Turner et al., 2020b).

To ensure a focus on highway emissions, we subtract prior estimates associated with non-highway sources from posterior BEACO₂N-STILT fluxes. Non-highway sources are small (~12%) in comparison with highway emissions, for the

190 pixels corresponding with the highway stretch analyzed in this study (Fig. 2, left), We assume the error in prior estimates of these sources to be an even smaller fraction of the total. For reference, a diel cycle of sector-specific, weekday prior emissions for the pixels analyzed in this study is shown in Fig. S3.

We estimate the BEACO₂N-STILT inversion to be precise to at least 30% for a line source. This estimate is based on the results of Turner <u>et al. (2016)</u> who used Observation System Simulation Experiments to demonstrate that with 7 days of observations at 30 sites a 45 tC/hr line source could be constrained to 15 t C/hr. However, this paper also demonstrated that error in the posterior decreased as results were averaged over a longer period of time, <u>Here</u> we are using 18 months, rather than 7 days of observations, we expect and observe better precision than 30%.

4

2.3 PeMS-EMFAC - derived CO₂ Emissions Estimates

Formatted: Font: 10 pt

Formatted: Line spacing: 1.5 lines

Deleted: (Delaria et al., 2021; Kim, Shusterman, Lieschke, Newman, & Cohen, 2018; Shusterman et al., 2016). The BEACO₂N network includes more than 70 locations in the SF Bay Area, spaced at ~2 km, and CO₂ measurements at individual sites have been shown to be accurate to 1.6 ppm

Formatted: Font: 10 pt

Formatted:	Font:	10 pt	

Formatted: Font: 10 pt

Formatted: Font: 10 pt

Deleted: sites actively reporting data

Formatted: Font: 10 pt

Formatted: Font: 10 pt

Deleted: To infer CO2 emissions from within the BEACO2N footprint, we use the Stochastic-Time Inverted Lagrangian Transport (STILT) model, coupled with a Bayesian inversion as described in detail in Turner et al 2020a (Turner, Kim, et al., 2020). Briefly, we use meteorology from NOAA's HRRR product at 3 km resolution to calculate footprints from each hour at each site, weighted by a priori CO2 emissions. The overall region of influence, the network footprint, as defined by a contour representing 40% of the CO2 influence is shown in Figure S1 (left). We construct a spatially gridded prior emissions inventory using point sources provided by the Bay Area Air Quality Management District (2011), home heating emissions as reported by BAAQMD (2011) and distributed spatially according to population density, on-road emissions from the HIghresolution Fuel Inventory for Vehicle Emissions (McDonald et al., 2014) varying by hour of week and scaled by year using fuel sales data, and a biogenic inventory derived using Solar Induced Fluorescence (SIF) Satellite data (Turner, Köhler, et al., 2020).

Formatted: Font: 10 pt

Formatted: Line spacing: 1.5 lines
Deleted: .
Deleted: that
Formatted: Font: 10 pt
Deleted: (2016) (Turner et al., 2016)
Formatted: Font: 10 pt
Deleted: , and because
Formatted: Font: 10 pt
Deleted: that this.
Formatted: Font: 10 pt

	Total hourly vehicle flow, truck (HDV) percent, and speed, were retrieved from http://pems.dot.ca.gov for January
	- June 2018-2020. There are ~1800 traffic counting stations hosted by the Caltrans Performance Measurement System
230	(PeMS) in the Bay Area, including more than 400 sites (Fig. S2) within the 2020 footprint of the BEACO2N, as described in
	Turner et al. (2020a), These stations count vehicle flow using magnetic loops imbedded in roadways and estimate HDV
	fraction using calculated vehicle speed and assumptions about vehicle length (Kwonvet al., 2003). For hours during which
	fewer than 50% of measurements were reported, we fill in total speed and light duty vehicle (LDV) flow gaps by using linear
	fits to nearest neighbor sites and gaps in HDV flow using hour-of-day- and weekend/weekday-specific median ratios
225	

235 between neighboring sites. We find that using this imputation method, mean absolute errors in speed are 5-10 km h⁻¹, in LDV flow are 500 vehicles h⁻¹, and in HDV flow are 50 vehicles / hour. (See Fig. S4.)

We calculate both LDV and HDV vkm for each highway segment during each hour, using downloaded flow data at each sensor location and segment lengths obtained from the PeMS database. For highway segments within the BEACO₂N footprint, vkm are summed to obtain regional highway HDV and LDV vkm for every hour. Figure <u>\$2</u>(left) shows the extent of the PeMS network in comparison to the BEACO₂N-STILT footprint, as well as total HDV vkm and LDV vkm.

Vehicle fuel efficiency is dependent on both fleet composition and vehicle speed. We calculate an emissions rate at \checkmark each location by combining speed and the HDV percentage with fuel efficiency estimates provided by the California Air Resources Board's Emissions FACtor Model (EMFAC2017). The EMFAC2017 model provides yearly fuel efficiency estimates for the Bay Area for 41 vehicle classes as a function of speed. We group these 41 vehicle types into the categories

245 LDV or HDV. (Table 55) PeMS's vehicle-type classification system is length based, assuming that LDV have a median length of 3.7 m and HDV a median length of 18.3 m (Kwon et al., 2003). As a result, we group most light duty trucks into the LDV category, To find speed-dependent emissions rate values for the LDV and HDV groups, we find a vkm-weighted mean of emissions rates across all vehicle-classes within a group at a given speed

 $er_{speed,group} = \frac{\sum_{i=1}^{n} vkm_{i,speed}er_{i,speed}}{\sum_{i=1}^{n} vkm_{i,speed}} (1)$

250 where *i* is a vehicle class. From this, we generate LDV and HDV emissions rates at 8.02 km h⁻¹ (5 mph) intervals. (See Fig. S6.) EMFAC does not provide data for several LDV vehicle classes at and above 96.8 km h⁻¹ (60 mph). To fill in this gap, we estimate emissions rates for the LDV group by using emissions rate to speed slopes (g CO₂ vkm⁻¹ km h⁻¹) for high speeds (88-145 km h⁻¹), using data from Davis et al. (2021).

We calculate emissions rates (g CO₂ / vkm) for each (< 1km) road segment between PeMS sensors at a moment in 255 time

 $er(t, seg) = \frac{vkm_{LDV}(t, seg)er_{LDV}(t, seg) + vkm_{tHDV}(t, seg)er_{tHDV}(t, seg)}{vkm_{LDV}(t, seg) + vkm_{HDV}(t, seg)}, (2)$

where emissions rates for cars and trucks are found via spline fit between reported speed for that segment and time with our curves for the emissions rates of each vehicle group. A fit is used rather than an individual bins, because of the sharp gradients that exist at low speeds for LDV. From the emissions rate for each (~1km) segment, we calculate an emissions rate for a stratch of high way including assumption for data taken and the provided of the sharp

protect for a stretch of highway including several segments to find total emissions rate (*er*) along a "stretch" over a period of time:

Formatted: Font: 10 pt

(Deleted: S1) within the 2020 footprint of the BEACO ₂ N, as described in Turner (2020a) (Turner, Kim, et al., 2020).
_(Formatted: Font: 10 pt
1	Deleted: , Varaiya, & Skabardonis,
,	Formatted: Font: 10 pt
//	Formatted: Indent: First line: 0.5", Line spacing: 1.5 lines
// (Deleted: 2
//	Formatted: Font: 10 pt, Font color: Text 1
///	Formatted: Font: 10 pt
///	Deleted: . In Figure 2 (right), we show
//	Formatted: Font: 10 pt
7,0	Formatted: Line spacing: 1.5 lines
/	Deleted: S1
//	Formatted: Font: 10 pt
///	Deleted:
///	Formatted: Font: 10 pt
///	Formatted: Font: 10 pt
//(Deleted: er(speed, group)
//(Formatted: Font: 10 pt
	Deleted: $\frac{\sum_{i=1}^{n} v k m_i e r_i}{\sum_{i=1}^{n} v k m_i}$,
// /	Formatted: Font: 10 pt
//	Deleted: kph (5 mph) intervals.
//	Formatted: Font: 10 pt
	Deleted: Because EMFAC does not provide data for several LDV vehicle classes at and above 96.8 kph (60 mph) we estimate emissions rates for the LDV group by using emissions rate to speed slopes (g CO ₂ vkm ⁻¹ kph ⁻¹) for high speeds (88-145 kph), using data from Argonne National Lab and EPA (Davis, Diegel, & Boundy, 2021). ^e
λ	Formatted: Font: 10 pt
λ	Deleted: then
-(Formatted: Font: 10 pt
(Formatted: Font: 10 pt
\nearrow	Formatted: Font: 10 pt
(Formatted: Line spacing: 1.5 lines

-1	Formatted: Font: 10 pt
(Formatted: Font: 10 pt
\nearrow	Formatted: Font: 10 pt
(Formatted: Line spacing: 1.5 lines
(Formatted: Indent: First line: 0", Line spacing: 1.5 lines
-(Deleted: class.
-(Formatted: Font: 10 pt
(Formatted: Font: 10 pt
(Formatted: Font: 10 pt

ard stratch	∑all se	$egments(vkm_{LDV}(t,s)er_{LDV}(t,s)+vkm_{HDV}(t,seg)er_{HDV}(t,s))$	(3)
	_	$\sum_{all \ segments}(vkm_{LDV}(t,s)+vkm_{HDV}(t,s))$	5)

Total CO2 emissions rates for the highway stretch analyzed in this work are shown in Fig. 2 (right, bottom),

3 Results

To gain insight into the relative impacts of congestion and fleet composition, we calculate fleet-wide vehicle emission rates (gCO₂/vkm) using two different methods. For both methods, the Caltrans Performance Measurement System (PeMS) provides vehicle counts, speed and categorizes HDV vs. LDV (http://pems.dot.ca.gov). Using this data and estimates of fuel per km from the EMissions FACtor 2017 (EMFAC) Model, we calculate the CO₂ emissions per km for the average vehicle with hourly time resolution as described above. Second, we use the PeMS data in combination with g CO₂ per unit area derived from the BEACO₂N-STILT inversion system. We focus on the ~5 km stretch of Interstate-80 just north of the San Francisco-Oakland Bay Bridge (Fig. 2). Interstate 80 is an East-West Highway whose orientation in this stretch is mainly North-South, with eastbound lanes traveling north and westbound lanes traveling south. The road has 5 lanes in each 290 direction and is often subject to high congestion (vehicles traveling slower than the posted speed).

PeMS-EMFAC-derived emissions rates give us insight into (1) the expected variation in emissions rates across a typical day (Fig. 2) and (2) the relative impacts of congestion vs. HDV percentage as factors leading to this variation (Fig. S7). For example, while the west-bound segment experiences speeds significantly below free-flow during both morning and evening rush hours, the east-bound segment experiences significant congestion only during the evening. Because of a steep

295 gradient in LDV emission rates between 20 and 50 km h⁻¹ (Fig. S6), the west-bound congestion in this segment occurs at speeds that are more fuel efficient than free-flow. The overall variance in emissions rates over the whole stretch is significantly smaller than in either of the directions shown individually.

From PeMS-EMFAC-derived emissions factors, we predict a median diel cycle with emissions per km travelled ranging from ~247 to ~314 g CO₂ / vkm. For reference, if all vehicles were driving at the speed limit of 104.6 km h⁻¹ (65 300 mph) and the fleet mix was 6% HDV and 94% LDV, we calculate an emission rate of 265 g CO₂ / vkm. The range of predicted emissions are narrower on the weekend (238 to 276 g CO₂ / vkm), both because fewer HDV use the road and because there is a smaller range in speed.

Figure S7 shows the hourly variation in the relative contributions of LDV speed, HDV percentage, and HDV speed to the deviation in g CO₂ / vkm from the reference value of 265 g CO₂ / vkm. The solid line is the mean and the shaded
and envelope represents the day-to-day variance. In the morning and mid-day, HDV percentage and LDV speed have opposite impacts on g CO₂ / vkm, leading to small variations in g CO₂ / vkm over the day, despite substantial variations in the separate effects of speed and HDV %. During evening rush hour, low vehicle speeds result in higher emission rates, leading to large positive deviations. High day-to-day variance in vehicle speed contributes to high day-to-day variance in emission rates. At times near midnight, large, positive deviations are observed, mostly as a consequence of high HDV percentage, but

Formatted: Line spacing: 1.5 lines Deleted: Sull segments VRMLDV(L3) erLDV(L3) + VRMLDV(L3) erLDV(L3) Sall segments VRMLDV(L3) + VRMLDV(L3) Formatted: Font: 10 pt Formatted: Font: 10 pt Deleted: Figure 2b. Formatted: Font: 10 pt Formatted: Indent: First line: 0.5", Line spacing: 1.5 lines

-(Deleted: Figure 2).
-(Formatted: Font: 10 pt
-(Deleted: and slow speeds.
~(Formatted: Font: 10 pt
(Formatted: Line spacing: 1.5 lines
(Formatted: Font: 10 pt
(Deleted: Figure 2
(Formatted: Font: 10 pt
(Deleted: The
(Formatted: Font: 10 pt
~(Formatted: Font: 10 pt

Deleted: kph

Formatted: Font: 10 pt

Formatted: Font: 10 pt

also because traffic flows at rates higher than 104.6 kph, leading to higher emission rates. Night-to-night variance in HDV percentage is low, thus variance in nighttime predicted g CO_2 / vkm is small. HDV speed has little impact on g CO_2 / vkm.

We use CO₂ measurements from 50 BEACO₂N sites across the Bay Area, combined with the BEACO₂N-STILT inversion system to <u>assess</u> highway emissions from our stretch of interest. In Fig. 1, we show the location of BEACO₂N sites, the stretch of interest, and emissions estimates for this stretch. Note that the posterior emissions move substantially from prior emissions towards what is estimated from PeMS-EMFAC, particularly during evening rush hour, when the prior overestimates emissions by ~20%.

We compare BEACO2N-derived and PeMS-EMFAC-derived emissions rates (CO2/ vkm) and find remarkable
agreement. The PeMS-EMFAC-derived emissions rates range from 225-300 g CO2/ vkm and include effects of both fleet
composition and variation in speed. For BEACO2N, we use the total CO2 emissions from the inversion at times
corresponding to narrow bins of PeMS-EMFAC g CO2/ vkm. Figure 3 (left) shows an example of data selected at times with
with PeMS-EMFAC-derived fuel efficiency in the range 271.4-279 g CO2/ vkm. There is a range of emissions at each vkm
because of noise in the inversion, variation in speed and variation in fleet composition. The slope of a fit to the data in Fig. 3
(left) is an estimate of the emissions rate (equation 4), where CO2 emissions is defined as hourly emissions summed over

BEACO₂N pixels corresponding to our highway stretch of interest (Fig. 2)

$er(g CO_2/vkm) = \frac{CO_2 \ emissons}{vkm}$ (4)

Using 18 months of data for weekdays between 4 am and 10 pm, we compare PeMS-EMFAC-derived and BEACO₂N-derived CO₂ / vkm (Fig. 3, right). These hours were chosen, because they represent the hours for which we

- 335 expect traffic emissions to be substantially larger than emissions from other sources in our area of interest (See Fig. S3), Fitting to a line forced through the origin, emissions rates found via the BEACO₂N inversion are within 3% (0.97 +/- 0.01) of those predicted using PeMS-EMFAC traffic counts. A more complete description of this fitting and error calculation process can be found in Text S8 and a comparison to results from applying this method to the prior can be found in S9. Using the definition of limit of detection as three times our uncertainty, we calculate that we would be able to detect an 11%.
- 340 change in individual points (representing bins of fuel efficiency from a combination of HDV percent and speed) and a 3% change in the slope. Because 18 months of data was required to reach this level of certainty, if we assume the 2.3-3.8% per year decrease in emission rate found by Kim, et al. 2021, we should be able to detect a change in overall fuel efficiency with three full years of BEACO₂N-STILT output.

We also consider how emissions rates compare, throughout the day (Fig. 4, top). During the evening, PeMSEMFAC-derived and BEACO₂N-derived emission rates are in good agreement. The BEACO₂N g CO₂/vkm increases from 256 g CO₂ / vkm before rush hour (2 pm) to 324 g CO₂/ vkm during peak rush hour (5 pm). Likewise, the PeMS-EMFAC-derived CO₂/vkm increases from 256 CO₂ / vkm to 320 CO₂ / vkm over the same time period. The BEACO₂N prior has a slightly larger increase in emission rate, over this period (256 g CO₂/vkm at 2PM to 361 g CO₂/vkm, at 5PM). In contrast, during the morning rush hours, we see less agreement between PeMS-EMFAC-derived and BEACO₂N-derived emission

1		
	Formatted	[36]
b	Formatted	([37])
Ζ,	Deleted: estimate	
6	Formatted	([38])
Ļ	Deleted: Figure	([50])
er. 	Formatted	([39])
	Deleted: estimates	([39])
	Formatted	
	Deleted: during which	[40]
-	Formatted	
	Formatted	([41])
1		[42]
Ŋ	Deleted: Figure	$ \longrightarrow $
IJ	Formatted	([43])
9	Deleted: Figure 3).	$ \longrightarrow $
h	Formatted	[44]
7	Deleted: CO ₂ /vkm =)
h	Formatted	[45]
Ų	Deleted: ,	
2	Formatted	[46]
/	Deleted: all hours	
_	Formatted	[47]
	Deleted: Figure 3, right).	
_	Formatted	[48])
	Deleted: Because eight of the nine points corresponding	t [49]
l,	Formatted	([50])
1	Deleted: fit, we estimate that	
0	Formatted	
1		[51]
	Deleted: BEACO2N system	([51])
	Deleted: BEACO ₂ N system	\square
	Formatted	[51] [52]
<	Formatted Deleted: a	[52]
	Formatted Deleted: a Formatted	\square
	Formatted Deleted: a Formatted Deleted: emission rates on	([52]) ([53])
	Formatted Deleted: a Formatted Deleted: emission rates on Formatted	[52]
	Formatted Deleted: a Formatted Deleted: emission rates on Formatted Deleted: order of 5%.	([52]) ([53]) ([54])
	Formatted Deleted: a Formatted Deleted: emission rates on Formatted Deleted: order of 5%. Formatted	([52]) ([53])
	Formatted Deleted: a Formatted Deleted: emission rates on Formatted Deleted: order of 5%. Formatted Deleted: agree	([52]) ([53]) ([54]) ([55])
	Formatted Deleted: a Formatted Deleted: emission rates on Formatted Deleted: order of 5%. Formatted Deleted: agree Formatted	([52]) ([53]) ([54])
	Formatted Deleted: a Formatted Deleted: emission rates on Formatted Deleted: order of 5%. Formatted Deleted: agree Formatted Deleted: Figure	([52]) ([53]) ([54]) ([55]) ([56])
	Formatted Deleted: a Formatted Deleted: emission rates on Formatted Deleted: order of 5%. Formatted Deleted: agree Formatted Deleted: Figure Formatted Formatted Formatted Formatted	([52]) ([53]) ([54]) ([55])
	Formatted Deleted: a Formatted Deleted: emission rates on Formatted Deleted: order of 5%. Formatted Deleted: agree Formatted Deleted: Figure Formatted Formatted Formatted Formatted	([52]) ([53]) ([54]) ([55]) ([56])
	Formatted Deleted: a Formatted Deleted: emission rates on Formatted Deleted: order of 5%. Formatted Deleted: agree Formatted Deleted: Figure Formatted Formatted Formatted Formatted	(
	Formatted Deleted: a Formatted Deleted: emission rates on Formatted Deleted: order of 5%. Formatted Deleted: agree Formatted Deleted: Figure Formatted Deleted: higher Formatted Deleted: higher	([53] ([54] ([55] ([55]) ([55])
	Formatted Deleted: a Formatted Deleted: emission rates on Formatted Deleted: order of 5%. Formatted Deleted: agree Formatted Deleted: Figure Formatted Deleted: Figure Formatted Deleted: higher	([52] ([53] ([54] ([55]) ([55]) ([55]) ([55])
	Formatted Deleted: a Formatted Deleted: emission rates on Formatted Deleted: order of 5%. Formatted Deleted: agree Formatted Deleted: Figure Formatted Deleted: higher Formatted Deleted: higher	([52] ([53] ([54] ([55]) ([55]) ([55]) ([55])
	Formatted Deleted: a Formatted Deleted: emission rates on Formatted Deleted: order of 5%. Formatted Deleted: agree Formatted Deleted: rigure Formatted Deleted: higher Formatted Deleted: higher Formatted Deleted: rates	([53] ([54] ([54] ([55] ([55] ([55] ([57] ([58])
	Formatted Deleted: a Formatted Deleted: emission rates on Formatted Deleted: order of 5%. Formatted Deleted: agree Formatted Deleted: rigure Formatted Deleted: higher Formatted Deleted: higher Formatted Deleted: rates Formatted	
	Formatted Deleted: a Formatted Deleted: emission rates on Formatted Deleted: order of 5%. Formatted Deleted: agree Formatted Deleted: Figure Formatted Deleted: higher Formatted Deleted: rates Formatted Deleted: rates Formatted	

- 395 rate estimates. The BEACO₂N inversion is similar to the PeMS-EMFAC estimate at 5 am local time (280 g CO₂ / vkm) and then the BEACO₂N estimate increases over the morning rush hour to 330 g CO₂ / vkm at 8 am. This behavior is different than either the BEACO₂N prior (175 at 5 am and 275 at 8 am) or the PeMS-EMFAC calculation which decreases over this period (275 at 5 am and 250 at 8 am).
- The discrepancy in the morning between emissions derived from PeMS-EMFAC and BEACO₂N can potentially be reconciled by congestion. There is a non-linear relationship between vehicle speed and the rate of emissions. As such, congestion involving non-constant speeds can result in higher emissions than would be estimated using the average vehicle speed. This can be seen from a simple example. Consider two cases: 1) a LDV travelling at a constant 50 km h⁻¹ for one hour and 2) a LDV traveling at 100 km h⁻¹ for 20 minutes and 25 km h⁻¹ for 40 minutes. Both vehicles travel 50 km in one hour and therefore have the same average speed. However, the emissions rate is 461.5 g CO₂/vkm at 25 km h⁻¹ 195 g
- 405 CO₂/vkm at 50 km h⁻¹ and 221 g CO₂/vkm at 100 km h⁻¹. Using these emission rates, the vehicle in the first case would emit 9.75 kg CO₂ whereas the vehicle with the variable speed in the second case would emit 15 kg CO₂.

<u>Contrasting the speeds (Fig. 4 bottom, right) during these two periods, we see that while both show a bi-modal</u> speed distribution, a greater fraction of morning speeds fall into the 40-100 kph range, whereas a greater fraction of evening speeds are < 40 km h⁻¹ or > 100 km h⁻¹. We show in Fig. S10, emission rate estimates based on hourly averaged speeds

410 between 0-40 km h⁻¹ and 100-140 km h⁻¹ (more common in evening rush hour) are likely an upper bound on possible emission rates corresponding to those hourly averaged speeds, whereas emission rate estimates based on hourly averaged speeds between 40-100 km h⁻¹ (more common in morning rush hour) likely represent a lower bound of emissions. The predicted range in emission rate resulting from non-constant speeds, combined with a larger HDV % in the morning (Fig. 4 bottom, right), is large enough to explain the mismatch observed during morning rush hour.

415 4 Discussion

Strategic reduction of emissions from transportation is important to both reducing total GHG emissions and improving AQ. To make informed decisions that reduce GHGs and exposure to poor AQ, policy makers need to know (1) how much is being emitted, (2) location and timing of emissions, and (3) the relative impact of various sub-sector processes (vkm, fleet composition, congestion).

420

To effectively capture emissions from sub-sector processes, models are also reliant on emissions factor models, such as the EMFAC2017 emissions model used in this paper. <u>While our BEACO₂N-STILT based estimates largely agree</u> with the EMFAC2017 emissions model for CO₂, tracking on-road changes in emission factors will be especially important as the impacts of congestion and fleet composition evolve rapidly making timely updates essential to creating spatially accurate inventories. For example, the EMFAC model predicts an 18% decrease in overall CO₂ emission rates by 2030,

425 resulting from the improved fuel efficiency of combustion engine vehicles and a transition to hybrid and EV (~6.8% of LDV vkm and ~6% of HDV vkm are expected to be <u>traveled</u> by EV by 2030). While the increased share of hybrid and EV should

Formatted: Font: 10 pt

Deleted:	
Deleted: kph	
Formatted: Font: 10 pt	
Formatted: Font: 10 pt	
Deleted: kph	
Deleted: kph	
Formatted: Font: 10 pt	
Deleted: 1	
Formatted: Font: 10 pt	
Formatted: Font: 10 pt	
Deleted: kph	
Formatted: Font: 10 pt	
Deleted: kph	
Deleted: kph	
Formatted: Font: 10 pt	
Formatted: Font: 10 pt	
Formatted: Line spacing: 1.5 lines	

Formatted: Font: 10 pt

Deleted: While our measurements largely agreed with the EMFAC2017 emissions model for CO₂, plume-based emission factor measurements of co-emitted pollutants (CO, NOX, PM2.5, BC, NMHC) show various emissions factor models to systematically underestimate emissions factors (Bishop, 2021), fail to capture spatial heterogeneity in these factors due to fleet composition (age and compliance with control technologies) for PM (Haugen & Bishop, 2018; Park, Vijayan, Mara, & Herner, 2016) and Black Carbon (Preble, Cados, Harley, & Kirchstetter, 2018), or fail to capture the impact of temperature on emissions factors.

Formatted: Font: 10 pt

Deleted: in interactive ways	
Formatted: Font: 10 pt	
Deleted: travelled	
Formatted: Font: 10 pt	

work to decrease the impact of congestion, a projected increase in total congestion and congested-vkm share by HDV (Texas A&M Transportation Institute, 2019) is likely to work against that trend, making the overall result difficult to predict.

To our knowledge, this paper represents the first demonstration that a high-density <u>atmospheric observing</u> network can both diagnose and quantify relative contributions of sub-sector processes at the neighborhood scale. We demonstrate that <u>the BEACO₂N</u> network (~2 km spacing) of low-cost CO₂ sensors, can be used to quantify emission rates at a specific location (~5 km stretch) and by time of day. We show that on the highway stretch, activity-based emissions estimates that account for speed and HDV % match the inference from atmospheric measurements to within 3%. Finally, we demonstrate

455 that the BEACO₂N-STILT system detects <u>daily changes in fuel efficiency that range from 200-300 g CO₂ / vkm and this system would be capable of detecting fleet-wide changes in fuel efficiency in ~3 years,</u>

5 Outlook

- 460 In this work, we have demonstrated that the BEACO₂N-STILT system was able to infer emission rates from vehicles along a specific stretch of highway. To understand the extent to which this method can be applied to other contexts, future work should investigate the extent to which various elements of the BEACO₂N-STILT system, including measurement density, error in meteorology used to calculate STILT trajectories, and the quality of the prior, impact the ability of similar systems to estimate emissions.
- For example, it is possible that the mismatch we observe during the morning rush hour may be due to a larger relative meteorological model error during the morning as compared to the afternoon and early evening in which the boundary layer is relatively well mixed. Because a highly mixed boundary layer is important for minimizing discrepancies between particle trajectories in the STILT model and real transport (Lin et al., 2003), inversions typically use only measurements taken during the afternoon, (Lauvaux et al., 2016; Nathan et al., 2019; Lauvaux et al., 2020) when the
- 470 boundary layer is relatively well mixed. However, as discussed by Martin et al. (2019), the impacts of meteorological mismatch during the morning may be offset by stronger signal, and future work should explore the extent to which averaging results over long time periods or strategic filtering of meteorological mismatches can combat emissions error.

Beyond further exploration of the elements influencing the sensitivity and precision of the BEACO₂N-STILT system, because each BEACO₂N node measures CO, NO_x, and PM_{2.5} in addition to CO₂ (Kim et al., 2018), the method

- 475 presented in this paper has the potential to shed light subsector processes impacting emission factors of these co-emitted species. This is salient because plume-based emission factor measurements of co-emitted pollutants show various emissions factor models systematically underestimate emissions (Bishop, 2021), fail to capture spatial heterogeneity in these factors due to fleet composition (age and compliance with control technologies) for PM (Haugen et al., 2018; Park, et al., 2016) and Black Carbon (Preble et al., 2018), or fail to capture the impact of temperature on emissions factors.
- 480 Applying these methods across a broader spatial area and to other species (PM_{2.5}, NO_x, CO) should yield information of interest to both scientists and policy makers by:

Deleted: surface

~~(Formatted: Font: 10 pt
(Deleted: using atmospheric data.
\mathcal{A}	Formatted: Font: 10 pt
\mathcal{I}	Deleted: atmospheric measurements, specifically a dense
)(Formatted: Font: 10 pt
(Deleted: that these variations are accurate to within approximately 5%
(Formatted: Font: 10 pt
\geq	Formatted: Font: 10 pt, Font color: Auto

Formatted: Font: 10 pt

Formatted: Indent: First line: 0.5", Line spacing: 1.5 lines

1. Revealing spatial and temporal trends in <u>emission</u> rates and emission factors across an urban area and quantifying the	Deleted: CO ₂ emissions
contributions of congestion, fleet composition, or other factors to spatial variations.	Deleted: spatially
2. Identifying and diagnosing the causes of traffic-related AQ hotspots that contribute to exposure inequities.	
 3. Tracking trends in the above over periods of years to decades. 	Deleted: <#>Characterizing emissions rates and emissions factors as a function of location, congestion, fleet composition, or meteorology.
Author Contributions:	Formatted: Font: 10 pt
HLF derived CO2 emissions from traffic data, conceived of project design, wrote manuscript, collected CO2 data. AJT	Formatted: Line spacing: 1.5 lines
created and ran CO2 inversion code. HLF, JK, KC, ERD, CN, PW collected CO2 data. RCC gave feedback on project design,	Deleted: ED
95 assisted in writing manuscript.	Formatted: Font: 10 pt
Competing Interest Statement: We have no competing interests to disclose.	
Acknowledgments: HLF was supported by NSF GRFP fellowship and Microsoft Research Internship. Thanks to K. Lauter	
00 and MSR Urban Innovation Group for support in thinking through PeMS data acquisition. AJT was supported as a Miller	
Fellow with the Miller Institute for Basic Research in Science at UC Berkeley. This research was funded by grants from the	
Koret Foundation and University of California, Berkeley. This research used the Savio computational cluster resource	
provided by the Berkeley Research Computing program at the University of California, Berkeley (supported by the UC	
Berkeley Chancellor, Vice Chancellor for Research, and Chief Information Officer). Thanks to HSK for reading through and	
05 offering organizational suggestions on the manuscript.	
Data Availability: The CO2 data used for this study are publicly available at http://beacon.berkeley.edu (Cohen Research,	Deleted: avail- able
2021). Raw data can be given upon request. The traffic data used for this study is publicly available at	
https://pems.dot.ca.gov/.	
References	Formatted: Line spacing: 1.5 lines
10 Apte, J. S., Messier, K. P., Gani, S., Brauer, M., Kirchstetter, T. W., Lunden, M. M., Marshall, J.D., Portier, C.J.,	Formatted: Font: 10 pt
Vermeulen, R.C.H., Hamburg, S. P, High-Resolution Air Pollution Mapping with Google Street View Cars: Exploiting	Deleted:
Big Data. Environmental Science and Technology, 51(12), 6999–7008. https://doi.org/10.1021/acs.est.7b00891, 2017	Deleted: (2017).
Bishop, G. A, Does California's EMFAC2017 vehicle emissions model underpredict California light-duty gasoline vehicle	Deleted: (2021).
NOx emissions? Journal of the Air and Waste Management Association, 71(5), 597–606.	
15 https://doi.org/10.1080/10962247.2020.1869121, <u>2021.</u>	
Boswell, M. R., & Madilyn Jacobson, A. R. 2019 Report on the State of Climate Action Plans in California.	Deleted: (2019).
https://ww2.arb.ca.gov/sites/default/files/2020-03/17RD033.pdf, last accessed: January 12, 2022, 2019.	Deleted: FINAL REPORT Principal Investigator, (17).

530	California Air Resources Board 2018 PROGRESS REPORT: California's Sustainable Communities and Climate Protection		Deleted: (2018).
	Act, (November), 96. https://ww2.arb.ca.gov/sites/default/files/2018-	(Deleted: Retrieved from
	11/Final2018Report SB150 112618 02 Report.pdf, last accessed: January 12, 2022	(Deleted: legislatively-mandated-reports
	Caubel, J. J., Cados, T. E., Preble, C. V., & Kirchstetter, T. W, A Distributed Network of 100 Black Carbon Sensors for 100		Deleted: (2019).
	Days of Air Quality Monitoring in West Oakland, California. Environmental Science and Technology, 53(13), 7564-		
535	7573. https://doi.org/10.1021/acs.est.9b00282, 2019.		
	Davis, S. C., Diegel, S. W., & Boundy, R. G. Transportation Energy Data Book, Edition 29. Energy.	(Deleted: (2021).
	https://tedb.ornl.gov/wp-content/uploads/2021/02/TEDB_Ed_39.pdf, last accessed January 12, 2022		
	City of Oakland: Oakland Equitable Climate Action Plan. https://cao-94612.s3.amazonaws.com/documents/Oakland-ECAP-		Deleted: Daw, T. (2020). Oakland EQUITABLE CLIMATE
	07-24.pdf, last accessed January 12, 2022.	l	ACTION PLAN, (July).
540	Delaria, E.R., Kim, J., Fitzmaurice, H.L., Newman, C., Wooldridge, P.J., Worthington, K. and Cohen, R.C., The Berkeley	(Deleted:
	Environmental Air-quality and CO 2 Network: field calibrations of sensor temperature dependence and assessment of	\sum	Deleted:
	network scale CO 2 accuracy, Atmospheric Measurement Techniques, 14(8), pp.5487-5500.	$\langle \rangle$	Deleted:
	https://doi.org/10.5194/amt-14-5487-2021, 2021	Y/////	Deleted: ., &
	Gately, C. K., & Hutyra, L. R, Large Uncertainties in Urban-Scale Carbon Emissions. Journal of Geophysical Research:	////	Deleted:
545	Atmospheres, 122(20), 11,242-11,260. https://doi.org/10.1002/2017JD027359, 2017a.		Deleted: . (2021).
545	· · · · · · · · · · · · · · · · · · ·	//[Formatted: Line spacing: 1.5 lines
	Gately, C. K., Hutyra, L. R., Peterson, S., & Sue Wing, I Urban emissions hotspots: Quantifying vehicle congestion and air		Deleted: Network
	pollution using mobile phone GPS data. Environmental Pollution, 229, 496–504.	\mathbb{N}	Deleted: , (May), 1–30. Deleted: (2017).
	https://doi.org/10.1016/j.envpol.2017.05.091, 2017b.		Deleted: (2017).
	Gately, L. K., Hutyra, L. R., & Wing, I. S, Cities, traffic, and CO2: A multidecadal assessment of trends, drivers, and scaling	Y	Deleted: (2017).
550	relationships. Proceedings of the National Academy of Sciences of the United States of America, 112(16), 4999-5004.		Deleted: Conor
	https://doi.org/10.1073/pnas.1421723112, 2015.		Deleted: (2015).
	Gurney, K.R., Liang, J., Roest, G., Song, Y., Mueller, K., & Lauvaux, T., Under-reporting of greenhouse gas emissions in		Moved (insertion) [1]
	U.S. cities. Nature Communications, 12(1), 1-7. https://doi.org/10.1038/s41467-020-20871-0, 2021.	(Deleted: Kevin
	Gurney, K. R., Razlivanov, I., Song, Y., Zhou, Y., Benes, B., & Abdul-Massih, M. Quantification of fossil fuel CO2	(Formatted: Line spacing: 1.5 lines
555	emissions on the building/street scale for a large U.S. City. Environmental Science and Technology, 46(21), 12194-	(Deleted: (2012).
	12202. https://doi.org/10.1021/es3011282, 2012.		
	Haugen, M. J., & Bishop, G. A. Long-Term Fuel-Specific NOx and Particle Emission Trends for In-Use Heavy-Duty		Deleted: Gurney, Kevin Robert, Liang, J., Roest, G., Song, Y.,
	Vehicles in California. Environmental Science and Technology, 52(10), 6070-6076. research-article.		Mueller, K., & Lauvaux, T. (2021). Moved up [1]: Under-reporting of greenhouse gas emissions in
	https://doi.org/10.1021/acs.est.8b00621, 2018.		U.S. cities. Nature Communications, 12(1), 1-7.
560	IPCC. (2014). Climate Change 2014 Part A: Global and Sectoral Aspects. Climate Change 2014: Impacts, Adaptation, and		https://doi.org/10.1038/s41467-020-20871-0
1	Vulnerability. Part A: Global and Sectoral Aspects. Contribution of Working Group II to the Fifth Assessment Report		Deleted: 1 Deleted: (2018).
1	of the Intergovernmental Panel on Climate Change.		Deleted: Retrieved from papers2://publication/uuid/B8BF5043-
	Kim, J., Shusterman, A. A., Lieschke, K. J., Newman, C., & Cohen, R. C. The Berkeley Atmospheric CO2 Observation		C873-4AFD-97F9-A630782E590D
	Kin, s. ondoernaal, A. A., Desenke, K. s., rewinan, e., & concil, K. Cy The Derectly Autospheric CO2 Observation	(Deleted: (2018).

1	Network: Field calibration and evaluation of low-cost air quality sensors. Atmospheric Measurement Techniques,	
	11(4), 1937–1946. https://doi.org/10.5194/amt-11-1937-2018, 2018.	
	Kim, J. Turner, A.J., Fitzmaurice, H.L., Delaria, E.R., Newman, C., Wooldridge, P.J., Cohen, R.C. Observing annual trends	
	in vehicular CO2 emissions (submitted to Environmental Science and Technology), 2021.	
600	Kwon, J., Varaiya, P., & Skabardonis, A, Estimation of Truck Traffic Volume from Single Loop Detectors with Lane-to-	Deleted: (2003).
	Lane Speed Correlation. Transportation Research Record, 684(1856), 106–117. https://doi.org/10.3141/1856-11, 2003.	Formatted: Line spacing: 1.5 lines
	Lauvaux, T., Gurney, K. R., Miles, N. L., Davis, K. J., Richardson, S. J., Deng, A., Nathan, B. J., Oda, T. Wang, J. A.,	Deleted:
	Hutyra, L., Turnbull, J, Policy-relevant assessment of urban CO2emissions. Environmental Science and Technology,	Deleted: (2020).
	54(16), 10237-10245. https://doi.org/10.1021/acs.est.0c00343.2020.	
605	Lauvaux, T., Miles, N. L., Deng, A., Richardson, S. J., Cambaliza, M. O., Davis, K. J., Gaudet, B. Gurney, K. R., Huang, J.	Deleted:
	O'Keefe, D., Song, Y., Karion, A., Oda, T., Patarsuk, R., Razlivanov, I., Sarmiento, D., Shepson, P, Sweeney, C.	
	Turnbull, J. Wu, K, High-resolution atmospheric inversion of urban CO2 emissions during the dormant season of the	Deleted: (2016).
	Indianapolis flux experiment (INFLUX). Journal of Geophysical Research, 121(10), 5213-5236.	
	https://doi.org/10.1002/2015JD024473,2016.	
610	Lian, J., Bréon, F.M., Broquet, G., Lauvaux, T., Zheng, B., Ramonet, M., Xueref-Remy, I., Kotthaus, S., Haeffelin, M. and	
	Ciais, P., Sensitivity to the sources of uncertainties in the modeling of atmospheric CO 2 concentration within and in	
	the vicinity of Paris. Atmospheric Chemistry and Physics, 21(13), pp.10707-10726. https://doi.org/10.5194/acp-21-	
	<u>10707-2021, 2021.</u>	
	Martin, C.R., Zeng, N., Karion, A., Mueller, K., Ghosh, S., Lopez-Coto, I., Gurney, K.R., Oda, T., Prasad, K., Liu, Y. and	
615	Dickerson, R.R., Investigating sources of variability and error in simulations of carbon dioxide in an urban region.	
	Atmospheric environment, 199, pp.55-69. https://doi.org/10.1016/j.atmosenv.2018.11.013, 2019.	
	McDonald, B.C., McBride, Z.C., Martin, E.W. and Harley, R.A., High-resolution mapping of motor vehicle carbon dioxide ≼	Deleted: C. (University of
	emissions. Journal of Geophysical Research: Atmospheres, 119(9), pp.5283, 5298.	Deleted: . B
	https://doi.org/10.1002/2013JD021219, 2014.	Deleted: (University of
620	Moua, F. (2020). California Annual Fuel Outlet Report Results (CEC-A15), Energy Assessments Division, California	Deleted: . B Deleted: (University of C. B., &
	Energy Comission, https://www.energy.ca.gov/media/3874, last accessed January 13, 2022	Deleted: (University of C. B., &
	Nathan, B.J., Lauvaux, T., Turnbull, J.C., Richardson, S.J., Miles, N.L. and Gurney, K.R., Source sector attribution of CO2	Formatted: Line spacing: 1.5 lines
	emissions using an urban CO/CO2 Bayesian inversion system. Journal of Geophysical Research: Atmospheres,	Deleted: Research
	123(23), pp.13-611. https://doi.org/10.1029/2018JD029231, 2018.	Deleted: carbon dioxide emissions. <i>Journal of Geophysical Research: Atmospheres</i> , (May),
625	Newman, S., Xu, X., Gurney, K.R., Hsu, Y.K., Li, K.F., Jiang, X., Keeling, R., Feng, S., O'Keefe, D., Patarasuk, R. and	Deleted: -
	Wong, K.W., Toward consistency between trends in bottom-up CO 2 emissions and top-down atmospheric	Deleted: .Received
	measurements in the Los Angeles megacity. Atmospheric Chemistry and Physics, 16(6), pp.3843-3863.	Deleted: 2018
	https://doi.org/10.5194/acp-16-3843-2016, 2016.	Deleted:
1	Park, S. S., Vijayan, A., Mara, S. L., & Herner, J. D, Investigating the real-world emission characteristics of light-duty	Formatted: Line spacing: 1.5 lines
	Park, S. S., Vijayan, A., Mara, S. L., & Herner, J. D, Investigating the real-world emission characteristics of light-duty	Deleted: (2016).

	gasoline vehicles and their relationship to local socioeconomic conditions in three communities in Los Angeles,		
50	California. Journal of the Air and Waste Management Association, 66(10), 1031-1044.		
	https://doi.org/10.1080/10962247.2016.1197166,2016.		
	Preble, C. V., Cados, T. E., Harley, R. A., & Kirchstetter, T. W _{r.} In-Use Performance and Durability of Particle Filters on		Deleted: . (2018).
	Heavy-Duty Diesel Trucks. Environmental Science and Technology, 52(20), 11913-11921. research-article.		
	https://doi.org/10.1021/acs.est.8b02977, 2018.		
5	Shusterman, A. A., Teige, V. E., Turner, A. J., Newman, C., Kim, J., & Cohen, R. C., The Berkeley Atmospheric CO2		Deleted: . (2016).
	Observation Network: Initial evaluation. Atmospheric Chemistry and Physics, 16(21), 13449-13463.		
	https://doi.org/10.5194/acp-16-13449-2016, 2016.		
	Tessum, C. W., Paolella, D. A., Chambliss, S. E., Apte, J. S., Hill, J. D., & Marshall, J. D., PM2.5 polluters		Deleted: . (2021).
	disproportionately and systemically affect people of color in the United States. Science Advances, 7(18), 1-7.		
0	https://doi.org/10.1126/sciadv.abf4491 <u>, 2021.</u>		
	Texas A&M Transportation Institute. (2019). Urban Mobility Report 2019, 182.		
	https://static.tti.tamu.edu/tti.tamu.edu/documents/umr/archive/mobility-report-2019.pdf, last accessed January 13,		Deleted: Retrieved from http://web.minienm.nl/mob2015
	2022.	(Deleted: Mobiliteitsbeeld_2015
	Turner, A.J., Kim, J., Fitzmaurice, H., Newman, C., Worthington, K., Chan, K., <u>Wooldridge, P.J., Köehler, P., Frankenberg</u>		Deleted:
5	C. and Cohen, R.C. 2020, Observed impacts of COVID, 19 on urban CO 2 Emissions. Geophysical Research Letters,	(Deleted:
	47(22), p.e2020GL090037.Geophysical Research Letters, 47(22), 1–6. https://doi.org/10.1029/2020GL090037, 2020a.	\square	Deleted:
	Turner, A. J., Köhler, P., Magney, T. S., Frankenberg, C., Fung, I., & Cohen, R. C., A double peak in the seasonality of		Deleted: . (
	California's photosynthesis as observed from space. Biogeosciences, 17(2), 405–422. https://doi.org/10.5194/bg-17-	, /// X	Deleted:).
	405-2020, 2020b.	///	Deleted: Impacts
<u> </u>		- / \{	Deleted: -
)	Turner, A. J., Shusterman, A. A., McDonald, B. C., Teige, V., Harley, R. A., & Cohen, R. C., Network design for	$\langle V \rangle$	Deleted: Urban CO2
	quantifying urban CO2 emissions: Assessing trade-offs between precision and network density. Atmospheric		Deleted: . (2020).
	Chemistry and Physics, 16(21), 13465–13475. https://doi.org/10.5194/acp-16-13465-2016, 2016.	L (Deleted: . (2016).

Formatted: Font: +Headings (Times New Roman), Bold







Figure 1. Left: Map of the BEACO₂N Network shows all sites (blue dots) for which there are more than 4 weeks of data during the period analyzed (Jan-June 2018-2020). Red stars indicate location of PeMS monitors used in this study. Right (top): CO₂ values shown for a 'typical week' during time period 695 observed. Dark line represents the median value observed across all sites and times. Shaded envelope represents 1 sigma variance across the network and over the 2 year period. Right (bottom): CO₂ emissions on all highway pixels in the domain as derived from the inversion of BEACO₂N observations (blue), BEACO2N prior (black), and PeMS-EMFAC-based estimate (red). Shaded envelope shows variance in emissions during the 18-month analysis window.





Figure 3: Left: BEACO₂N-derived emissions vs. vkm for times corresponding to modeled emission rates of 271.4-279 g CO₂/ vkm. <u>Red points represent binned medians used in fitting</u>. Right: BEACO₂N-derived vs. PeMS-EMFAC derived emissions rates with uncertainty estimate. Black line shows fit weighted by variance: y = 0.97(.01)x. Grey envelope is 5% deviation from fit. Red line represents 1:1
 11.1



Figure 4: Top: Emissions rates by time of day on weekdays for PeMS-derived (red), BEACO2N-prior
 (blue), and BEACO₂N posterior (green). Bottom: Probability density functions of truck fraction (left) and speed (right) from weekday morning (5-9 am) and evening (4-8 pm) rush hour period on the segment of I-80 analyzed in the Results section. Y-axis represents the relative probability of HDV fraction (left) or averaged hourly speed (right). Speeds are from individual PeMS sensors, while truck fraction is aggregated over the whole stretch under consideration (both directions).

Page 1: [1] Style Definition	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Normal (Web)			
Page 1: [2] Style Definition	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Authors			
Page 1: [3] Style Definition	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Correspondence			
Page 1: [4] Style Definition	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Footer			
Page 1: [5] Style Definition	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Caption			
Page 1: [6] Style Definition	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Equation			
Page 1: [7] Style Definition	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Balloon Text			
Page 1: [8] Style Definition	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Affiliation			
Page 1: [9] Style Definition	Helen Fitzmaurice	1/23/22 11:14:00 AM	
List Paragraph			
Page 1: [10] Style Definition	Helen Fitzmaurice	1/23/22 11:14:00 AM	
MS title			
Page 1: [11] Style Definition	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Copernicus_Word_template	2		
Page 1: [12] Style Definition	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Name			
Page 1: [13] Style Definition	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Kontakt			
Page 1: [14] Style Definition	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Header			
Page 1: [15] Style Definition	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Bullets: Bulleted + Level: 1	+ Aligned at: 0.25"		
Page 1: [16] Style Definition	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Betreff			
Page 1: [17] Style Definition	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Heading 4			
Page 1: [18] Style Definition	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Heading 3			
Page 1: [19] Style Definition	Helen Fitzmaurice	1/23/22 11:14:00 AM	
II 1: 0		· ·	

I

Handing 7

Page 1: [21] Style Definit	tion Helen Fitz	maurice 1/23/22 11:14	:00 AM
		ft, Line spacing: single	
Page 1: [22] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 1: [23] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Line spacing: 1.5 lin	les		
Page 1: [24] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 1: [25] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 1: [25] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 1: [26] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 1: [26] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 1: [27] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 1: [28] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 1: [29] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 1: [30] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 1: [31] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 1: [32] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 1: [33] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 1: [34] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 1: [35] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Indent: First line: 0.5	5", Line spacing:	1.5 lines	
Page 7: [36] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 7: [37] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	

Font: 10 pt

I

Page 7: [39] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt	Relefi Fitzillaurice	1/23/22 11:14:00 AM	
-			
Page 7: [40] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 7: [41] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 7: [42] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 7: [43] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 7: [44] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 7: [45] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 7: [46] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 7: [47] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt	Helen Hizzindurice	1, 23, 22 11.14.00 AM	
Page 7: [48] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt	Relefi Fitzillaurice	1/23/22 11:14:00 AM	
-			
Page 7: [49] Deleted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
▼			
Page 7: [50] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 7: [50] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 7: [51] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt		_,,	
Page 7: [52] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt		1, 20, 22 1111100 AM	
Page 7: [53] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt	neien ritzillaurite	1/23/22 11.14:00 AM	
Page 7: [54] Formatted		1/22/22 11.14.00 AM	
Font: 10 pt	Helen Fitzmaurice	1/23/22 11:14:00 AM	
1			
Page 7: [55] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			

Page 7: [57] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 7: [58] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 7: [59] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 7: [60] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 7: [61] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
Font: 10 pt			
Page 7: [62] Formatted	Helen Fitzmaurice	1/23/22 11:14:00 AM	
F (10)			

Font: 10 pt

Supporting Information for

Assessing vehicle fuel efficiency using a dense network of CO₂ observations

Helen L. Fitzmaurice¹, Alexander J. Turner², Jinsol Kim¹, Katherine Chan³, Erin R. Delaria⁴, Catherine Newman⁴, Paul Wooldridge⁴, Ronald C. Cohen^{1,4}

1. Department of Earth and Planetary Science, University of California Berkeley, Berkeley, CA, 94720, **United States**

2. Department of Atmospheric Sciences, University of Washington, Seattle, WA, 98195, United States

3. Sacramento Metro Air Quality Management District, Sacramento, CA, 95814, United States

4. Department of Chemistry, University of California Berkeley, Berkeley, CA, 94720, United States

Contents of this file

<u>Text S1 - S10</u> Figures S1 - S4, S6, S7, S9, S10 Table S5

(Moved (insertion) [1]

Formatted: Font: 11 pt, Font color: Auto Formatted: Font: 11 pt, Not Bold, Font color: Auto

Introduction

In S1, we describe the time series of the number of BEACO₂N nodes reporting CO₂ during the from January through June for the years 2018-2020. In S2, we show the locations in the PeMS measurement network in the region of the SF Bay Area shown on the map, as well as an estimate of LDV and HDV VMT for a typical week in this domain. In S3, we describe the hourly BEACO₂N-STILT prior for the typical weekday for CO₂ emissions in the 1km pixels that encompass the highway stretch that is the focus of our analysis. The figure also shows vkm traveled for each hour on this stretch of highway. In S4, error analysis for PeMS values for speed, LDV and HDV volume is described. In S5, we list EMFAC2017 vehicle classes and indicate whether we have classified them as LDV or HDV based on estimated vehicle length. In S6, we show both LDV and HDV emissions rates as a function of speed. We also compare a piece-wise linear to a spline fit of these two curves. In S7, we show the diel cycle for contribution to total emissions by congestion and vehicle type as estimated using PeMS-EMFAC. In S8, we describe the calculation of uncertainty in emissions rates derived using the BEACO2N-STILT system. In S9, we derive emission rates from the BEACO₂N-STILT prior and discuss improvements of the posterior over the prior. In S10, we explore how non-constant speed may impact emissions rates for a given hourly average speed.

Text S1.

Throughout the period examined in this study, the number of BEACO₂N sensors reporting data varied from due to power or instrument failure.



Figure S3. Number of BEACO₂N sites reporting CO₂ data used in BEACO₂N-STILT inversion for January-June in 2018 (top) 2019 (middle) and 2020 (bottom)

Text S2.

The Caltrans Performance Measurement System Network consists of thousands of magnetic loop monitors imbedded in highways across the state of California (http://pems.dot.ca.gov). Each station consists of loop sensors in each lane that report hourly values for total vehicle flow, HDV percentage, and average speed. Using station locations, vehicle flow, and HDV percentage, hourly vkm can be calculated as outlined in the main text.



Figure S2: Left: Locations of Caltrans PeMS monitoring stations (black and red). The solid blue line marks the 40% contour of the BEACO₂N cumulative influence function during January – June 2020. Right: LDV vkm, HDV vkm estimated based on PeMS data. Moved (insertion) [2] Formatted: Font: 11 pt

Text S3.

We focus our analysis on the hours 4am – 10pm. During this period, emissions from traffic are much larger than all other sources in the pixels used in this analysis. From 11pm – 3am, total vkm and therefore emissions from traffic are low.



Figure S3: (top) Diel variation of total vkm (from PEMS observations) for the stretch of roadway indicated in Figure 2 for a typical weekday. (bottom) Prior estimates of emissions from biogenic sources (orange), vehicle emissions (blue), point sources and area sources (yellow).

Text S4.

We apply linear fits (for speed and LDV) and hourly ratios (for HDV) to nearest neighbors, second nearest neighbors, and third nearest neighbors to create modeled values for all times for which we have observations. Using these modeled values we estimate mean error and spread for all PeMS sites over the time period studied, finding that speed accurate to about 5km hr⁻¹, LDV/hr to ~300 vehicles and HDV to ~55 vehicles for the east and west directions of flow on I-80. Precision is much higher than these values as shown on the right.





Figure S4: Mean average error (left) and distribution of error (right) for modeled speed (top), LDV flow (middle), and HDV flow (bottom).

Text S5.

While EMFAC2017 provided speed-dependent emission rate estimates for 41 vehicle classes, PeMS characterizes vehicles in two categories based on length. In order to use EMFAC2017 emission rates in combination with PeMS traffic counts to estimate total emissions, we classify EMFAC2017 categories as LDV or HDV based on length.

EMFAC Vehicle Class	Grouping for this work	Moved (insertion) [3]
All Other Buses	<u>0</u>	Formatted: Font: 8 pt
<u>LDA</u>	<u>1</u>	Formatted Table
LDT1	<u>1</u>	Formatted: Font: 8 pt
LDT2	<u>1</u>	Formatted: Font: 8 pt
LHD1	<u>1</u>	Formatted: Font: 8 pt
LHD2	1	Formatted: Font: 8 pt
MCY		Formatted: Font: 8 pt
		Formatted: Font: 8 pt
MDV	<u>1</u>	Formatted: Font: 8 pt
<u>MH</u>	<u>Q</u>	Formatted: Font: 8 pt
Motor Coach	<u>0</u>	Formatted: Font: 8 pt
OBUS	<u>0</u>	Formatted: Font: 8 pt
<u>PTO</u>	<u>0</u>	Formatted: Font: 8 pt
<u>SBUS</u>	<u>0</u>	Formatted: Font: 8 pt
T6 Ag	<u>0</u>	Formatted: Font: 8 pt
T6 CAIRP heavy	<u>0</u>	Formatted: Font: 8 pt
T6 CAIRP small	<u>1</u>	Formatted: Font: 8 pt
T6 OOS heavy	<u>0</u>	Formatted: Font: 8 pt
T6 OOS small	<u>1</u>	Formatted: Font: 8 pt
		Formatted: Font: 8 pt
T6 Public	<u>0</u>	Formatted: Font: 8 pt
T6 instate construction heavy	<u>0</u>	Formatted: Font: 8 pt
T6 instate	<u>1</u>	Formatted: Font: 8 pt
construction small T6 instate heavy	<u>0</u>	Formatted: Font: 8 pt
T6 instate small	1	Formatted: Font: 8 pt
T6 utility	<u></u>	Formatted: Font: 8 pt
<u></u>	<u>0</u>	Formatted: Font: 8 pt
T7 Ag	<u>0</u>	Formatted: Font: 8 pt
T7 CAIRP	<u>0</u>	Formatted: Font: 8 pt
T7 CAIRP construction	<u>0</u>	Formatted: Font: 8 pt
T7 NNOOS	<u></u>	Formatted: Font: 8 pt
<u>T7 NOOS</u>	<u></u>	Formatted: Font: 8 pt
<u>17 POAK</u>	<u></u>	
PT SAN	<u>v</u>	Formatted: Font: 8 pt

T7 Public	<u>0</u>	Formatted: Font: 8 pt
<u>17.SWCV</u>	<u>0</u>	Formatted: Font: 8 pt
T7 Single	<u>0</u>	Formatted: Font: 8 pt
17 other port	<u>0</u>	Formatted: Font: 8 pt
T7 single construction	<u>0</u>	Formatted: Font: 8 pt
17 tractor	<u>0</u>	Formatted: Font: 8 pt
17 tractor	<u>0</u>	Formatted: Font: 8 pt
construction T7 utility	<u>0</u>	Formatted: Font: 8 pt
<u>T7IS</u>	<u></u>	Formatted: Font: 8 pt
UBUS	<u>0</u>	Formatted: Font: 8 pt
		Formatted: Font: Calibri
*******		aracterize as LDV or HDV based on length.
" "1" denotes LDV and "0"	denotes HDV.	Moved (insertion) [4]
		Formatted: Font: Calibri, 11 pt
		Formatted: Font: Calibri, 11 pt, Bold

Text S6.

As described in the main text, emission rates for LDV and HDV on each road segment between individual PeMS monitoring stations are computed hourly as a function hourly average speed. Here we show emission rates as a function of speed.

We also compare piece-wise linear fits to the spline fits used in this study. With the exception of emissions rates for LDV at speeds lower than 20 km h⁻¹, there is little difference between these fits. High uncertainty in emission rates at low hourly average speeds because of travel at non-constant speeds is likely to outweigh any difference between these fits (see Fig S7).



Figure S6: We show emission rates (g CO_2 / km) of different vehicle classes as a function of speed. (top and middle) Red lines indicate emission rates for individual vehicle classes as reported by EMFAC2017. Black lines indicate extrapolation using Oakridge National Lab data. Heavy blue lines indicate emission rates for LDV and HDV groups calculated by taking the vkm-weighted mean of emission rates for all vehicles within a group at a particular speed. (bottom) We compare piecewise-linear fits of this data to spline fits. Black lines indicate spline fit. Blue lines indicate piecewise-linear fits.

Text S7.

Figure S5 shows the hourly variation in the relative contributions of LDV speed, HDV percentage, and HDV speed to the deviation in CO_2 / vkm from the reference value of 265 g CO_2 / vkm . The solid line is the mean, and the shaded envelope represents the day-to-day variance. In the morning and mid-day, HDV percentage and LDV speed have opposite impacts on CO_2 / vkm , leading to smaller variations in CO_2 / vkm than the variations in the separate effects of speed and HDV %. During evening rush hour, low vehicle speeds result in higher emission rates, leading to large positive deviations. High day-to-day variance in vehicle speed contributes to high day-today variance in emission rates, shown as the envelope surrounding the solid line At times near midnight, large, positive deviations are observed, mostly as a consequence of high HDV percentage, but also because traffic flows at rates higher than 104.6 kph, leading to higher emission rates. Night-to-night variance in HDV percentage is low, thus variance in nighttime predicted CO_2 / vkm is small. HDV speed has little impact on CO_2 / vkm .



Moved (insertion) [5] Formatted: Font: 11 pt, Not Bold

Moved (insertion) [6] Formatted: Font: 11 pt, Not Bold Moved (insertion) [7] Formatted: Font: 11 pt, Not Bold

Figure S7: (Top) PeMS-EMFAC-derived emissions rate deviations from baseline of 6% of all vehicles HDV, and vehicle speed constant at 105 kph resulting from car speed, truck percentage, and truck speed for the average day on the week shown in Figure 3. (Bottom) Total deviation in emissions rate by hour of day. % Deviation (right axis) shows percent deviation for all curves from emissions rate of 6% HDV at 105 kph. For all plots, solid line represents median values and shaded area represents variance.

(Moved (insertion) [8]

Formatted: Font: 11 pt

Text S8.

Determination of Uncertainty in Emissions Rate Estimates

For the set of BEACO₂N emissions corresponding in time to the data in each 7.8 g CO_2 / vkm bin of PeMS-derived emissions rates, we find a $\mathsf{BEACO}_2N\text{-}\mathsf{derived}$ emissions rate estimate. To do this, we take all BEACO2N traffic emissions occurring simultaneously with the PeMS-derived emissions rates and further bin these points based on vkm, as shown in Figure 3. For each vkm bin, we then find the median emissions value and the variance of emissions values, σ_{i}^{2} . We assume the error in our estimate of the median emissions for each vkm bin to be $\delta ems = \frac{\sigma}{\sqrt{n}}.$ We then fit median emissions values to the line $ems = \frac{gCO_2}{u^3km_A}vkm,$

to find $\frac{gCO_2}{aukmt}$, using δems as weights in the MATLAB fitlm function, and take the reported SE in slope to be the error in our calculated $\frac{gCO_2}{aukmt}$.

Formatted: Font: 11 pt	
Deleted: for	
Formatted: Font: 11 pt	
Deleted: emission corresponding to	
Formatted: Font: 11 pt	
Deleted: timestamps given by	
Formatted: Font: 11 pt	
Deleted: in this bin	
Formatted: Font: 11 pt	
Formatted: Font: 11 pt, Not Bold	
Formatted: Font: 11 pt	
Formatted: Font: 11 pt, Not Bold	
Formatted: Font: 11 pt	
Formatted: Font: 11 pt, Not Bold	
Formatted: Font: 11 pt	
Formatted: Font: 11 pt, Not Bold	
Formatted: Font: 11 pt, Font color: Auto	
Formatted: Font: 11 pt	

Text S9.

The prior inventory was constructed to reflect vehicle type (LDV v. HDV) dependence on emissions, but not speed-dependence in emissions. In order to illustrate improvement of the posterior (Figure 3) over the prior, we repeat the analysis described in the main text to show emission rates calculated for the prior. Calculated emissions rates for the prior are nearly constant over a wide range (237.5 – 262.5 g CO_2 / vkm) of PeMS-EMFAC emission rates. Where they do vary, they are substantially different than those estimated in the posterior.



Figure S9:

Emission rate estimates calculated for the BEACO₂N-STILT prior in the same manner in which they were calculated for the posterior vs. PeMS-EMFAC emissions estimates with uncertainty estimate. Black line shows fit of to posterior (Fig 3) weighted by variance: y = 0.97(.01)x. Grey envelope is 5% deviation from fit. Red line represents 1:1 line. Moved up [2]: Left: Locations of Caltrans PeMS monitoring stations (black and red). The solid blue line marks the 40% contour of the BEACO₂ x cumulative influence function during January – June 2020.

(... [2])

Moved up [8]: (Top) PeMS-EMFAC-derived emissions rate deviations from baseline of 6% of all vehicles HDV, and vehicle speed constant at 105 kph resulting from car speed, truck percentage, and truck speed for the average day on the week shown in Figure 3. (Bottom) Total deviation in emissions rate by hour of day. % Deviation (right axis) shows percent deviation for all curves from emissions rate of 6% HDV at 105 kph.



Text S10.

While PeMS reports hourly averaged speeds for each sensing station, non-constant speeds due to congestion can result in range of possible emissions rates that can occur for a particular hourly averaged speed.



Figure: S10 The dark line indicates the emissions rate corresponding to driving the speed indicated on the x axis at a constant velocity. The shaded region represents *er* distribution resulting from vehicle travel at non-constant speeds. For each speed, we calculate all possible emissions rates (g CO_2 / vkm) that could be generated assuming that the vehicle fleet (here, 8% HDV as is common during AM rush hour) drives at 2 different speeds between 8 kph and 130 kph for the times required to result in the average speed represented on the x axis. The spread for each speed represents the 16th-84th percentiles of possible emissions rates.

	ormatted: Font: 11 pt
F	ormatted: Font: 11 pt
D	eleted: then
F	ormatted: Font: 11 pt
	loved up [1]: ¶ ext
co de Ti	loved up [5]: shows the hourly variation in the relative ontributions of LDV speed, HDV percentage, and HDV speed to the eviation in CO ₂ / vkm from the reference value of 265 g CO ₂ / vkm. he solid line is the mean, and the shaded envelope represents the ay-to-day variance.
re H	loved up [6]: During evening rush hour, low vehicle speeds esult in higher emission rates, leading to large positive deviations. igh day-to-day variance in vehicle speed contributes to high day- o-day variance in emission rates
D	eleted: ¶
M	loved up [4]: Breakdown of EMFAC vehicle classes we
F	ormatted: Font: 11 pt, Font color: Auto
D	eleted: S3. Figure S3
F	ormatted: Font: 11 pt
D	eleted: In the morning and mid-day, HDV percentage and [4]
D	eleted: .
F	ormatted: Font: 11 pt
F	ormatted: Font: 11 pt, Not Bold, Font color: Auto
F	ormatted: Font: 11 pt
M	loved up [7]: At times near midnight, large, positive deviations
F	ormatted: Font: Calibri, 11 pt, Not Bold
F	ormatted: Font: Calibri, 11 pt
F	ormatted: Font: Calibri, 11 pt
M	loved up [3]: EMFAC Vehicle Class
F	ormatted: Font: 8 pt
F	ormatted Table
F	ormatted: Font: 8 pt
E	ormatted: Font: 8 pt
5	

Formatted: Font: 11 pt

Formatted: Font: 8 pt
Formatted: Font: 8 pt
Formatted: Font: Calibri

Page 11: [1] Deleted	Helen Fitzmaurice	1/23/22 11:10:00 AM	
V			
Page 11: [2] Deleted	Helen Fitzmaurice	1/23/22 11:10:00 AM	
v			
Page 11: [3] Deleted	Helen Fitzmaurice	1/23/22 11:10:00 AM	
V			
Page 12: [4] Deleted	Helen Fitzmaurice	1/23/22 11:10:00 AM	
•			
Page 12: [5] Deleted	Helen Fitzmaurice	1/23/22 11:10:00 AM	
_		· ·	