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# Contrasting Summertime Trends in Vehicle Combustion Efficiency in Los Angeles, CA and Salt Lake City, UT

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emissions of an pointains from motor venicles. As a result, venicle neets are expected to progressively combust fuel more efficiently, with a declining ratio of carbon monoxide to carbon dioxide  $(CO/CO_2)$  in their emissions. We assess trends in traffic combustion efficiency in Los Angeles (LA) and Salt Lake City (SLC) by measuring changes in summertime on-road  $CO/CO_2$  between 2013 and 2021 using mobile observations. Our data show a reduction in  $CO/CO_2$  in LA, indicating an improvement in combustion efficiency that likely resulted from stringent regulation of CO emissions. In contrast, we observed an increase in  $CO/CO_2$  values in SLC. While



slower progress in SLC compared to LA may be partially due to a later adoption of vehicle emission regulations in Utah compared to California, differing driving conditions and fleet composition may also be playing a role. This is evidenced by increased  $CO/CO_2$  in LA during the COVID-19 pandemic, which led to faster driving speeds and changes to the fleet composition. Our results demonstrate the success of California's CO-reducing policy interventions and illustrate the impacts of traffic characteristics on vehicle combustion efficiency and air pollutant emissions.

**KEYWORDS:** cities, transportation, emissions, air quality, Los Angeles, Salt Lake City

#### 1. INTRODUCTION

Urban areas are hotspots for the emission of trace gases that are harmful to human health and the environment. Many of these air pollutants and greenhouse gases are emitted on road from vehicles. In cities, the transportation sector accounts for a large share of anthropogenic carbon monoxide (CO) and carbon dioxide  $(CO_2)$  emissions, since both gases are coemitted during the burning of petroleum-derived fuels in gasoline- and diesel-powered vehicles. In the United States, 38% of anthropogenic CO emissions<sup>1</sup> and 29% of  $CO_2$ emissions<sup>2</sup> originate from on-road mobile sources. CO, a criteria pollutant that results from incomplete or inefficient combustion, is hazardous to human health, a precursor to other harmful air pollutants such as ozone, and contributes to climate change. CO<sub>2</sub> is a potent greenhouse gas and the most critical determinant of climate change in this century due to the unprecedented magnitude of emissions from human activity and its long-term persistence in the atmosphere.<sup>3</sup>

Air pollutant regulation (i.e., the 1970 U.S. Clean Air Act) and technological advances (i.e., catalytic converters) have led to significant improvements in air quality and a decline in air pollution-related mortality in many cities.<sup>4–6</sup> Specifically, requirements for catalytic converters on new vehicles reduced the amount of CO and unburned hydrocarbons emitted from vehicles by oxidation reactions that convert them to  $CO_2$ . Newer vehicle models are more fuel-efficient, and CO emissions have decreased despite increases in the number of

vehicles on the road and vehicle miles traveled. Ambient CO mixing ratios have substantially declined in urban areas,<sup>7</sup> especially within jurisdictions that enforce strict emission controls, such as the State of California. In the San Francisco metropolitan area, for instance, CO mixing ratios measured in vehicles traveling on a major highway decreased from 9.7 ppmv to 0.5 ppmv (a 95% reduction) between 1980 and 2011.<sup>12</sup> In Los Angeles, the CO mixing ratio decreased from 20 ppmv in the 1960s to 0.4 ppmv in 2010, with an average decrease of 8% per year.<sup>9</sup> However, while urban CO emissions have declined, global CO<sub>2</sub> emissions from the on-road sector have continued to grow due to increasing transportation.<sup>13</sup> In contrast to technological advances that reduced CO, reducing CO<sub>2</sub> emissions requires higher fuel economy or fundamental changes to the vehicle fuel source (e.g., electric and hydrogen vehicles).

The ratio of  $CO/CO_2$  is a useful metric that is indicative of vehicle combustion efficiency. Combustion efficiency denotes how effectively the vehicle engine converts fuel into energy, and consequently the degree to which polluting byproducts

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(i.e., CO) are emitted in the process. Lower CO/CO<sub>2</sub> ratios indicate improvements in combustion efficiency as well as the effectiveness of catalytic converters equipped by gasoline vehicles in reducing CO emissions, and thus the ratio can be used to evaluate the success of efforts to reduce pollutant emissions from traffic. In cities, the CO/CO<sub>2</sub> ratio has generally been declining over time alongside reductions in CO emissions.<sup>14</sup> However, the trend varies by location, reflecting the stringency and timing of emission reduction policies.<sup>9</sup> As such, CO/CO<sub>2</sub> ratios tend to be higher in places that have less stringent pollution regulations and vehicle fleets with less efficient technology.<sup>15,16</sup>

In addition to serving as an indicator of combustion efficiency, the  $CO/CO_2$  ratio is also a useful tracer for studies aiming to quantify fossil fuel CO<sub>2</sub> signals based on atmospheric CO<sub>2</sub> measurements. This is because CO is a short-lived gas, with an atmospheric lifetime of 1-2 months, compared to CO<sub>2</sub>, which stays in the atmosphere for centuries. Additionally, CO is coemitted with CO<sub>2</sub> during incomplete combustion of fossil fuels. This makes the  $\rm CO/\rm CO_2$  ratio unique for fossil fuel CO<sub>2</sub> emissions when compared to other CO<sub>2</sub> sources such as biogenic fluxes, oceanic exchange and wildfires.<sup>15,17-19</sup> Observations of  $CO/CO_2$  ratios provide critical insight regarding CO<sub>2</sub> sources and for validating the success of emission reduction efforts. However, spatial and temporal variations in the CO/CO<sub>2</sub> ratio complicate its application as a tracer. Thus, changes to the  $CO/CO_2$  ratio over time need to be reassessed to determine the continued usefulness of this approach.

In this study, we investigate multiyear trends in the traffic combustion efficiency using observations of the on-road CO/ CO<sub>2</sub> ratio in greater Los Angeles (LA) and Salt Lake City (SLC), two major metropolitan areas in the western U.S. with air quality problems and contrasting vehicle emission control policies. Measurements were conducted in both cities during the summers of 2013 and 2019, and additionally in LA in 2020 and 2021 to capture changes in traffic related to the coronavirus-19 (COVID-19) pandemic. We measured onroad CO/CO<sub>2</sub> using mobile laboratories equipped with fast response, high precision trace gas analyzers. Alternative approaches for estimating on-road emissions rely on assumptions about the traffic fleet characteristics and driving conditions in order to extrapolate measurements made in emission testing laboratories (e.g., EPA MOVES) or on individual vehicles (e.g., portable emissions measurement systems). In contrast, our measurements represent the actual average combustion efficiency of the real-world vehicle fleet on interstate highways during daytime in these locations. We chose interstate highways to avoid the influence of non-vehicle sources, cold starts, and to keep our intercity comparisons free of biases related to road network design. Thus, our approach allows a direct assessment of the integrated impact of vehicle emission control policies, vehicle fleet dynamics, and traffic conditions on air quality near roadways. We hypothesized that LA would have a lower and more rapidly decreasing  $CO/CO_2$ ratio than SLC due to the earlier adoption of strict vehicle emission control policies in California compared to Utah.

#### 2. MATERIALS AND METHODS

**2.1. Study Areas.** We focus on two urban areas in the western U.S.: Los Angeles, CA and Salt Lake City, UT. The Los Angeles (LA) metropolitan area hosts 18 million residents across approximately 88,000 km<sup>2</sup> in Southern California, while

Salt Lake City (SLC) has 1.2 million residents in approximately 20,000 km<sup>2</sup>. Both locations have historically suffered from poor air quality due to anthropogenic pollutant emissions that get trapped by atmospheric temperature inversions and surrounding mountains.<sup>20-22</sup> Both LA and SLC are classified as maintenance areas for ambient CO levels due to former violations of air quality standards and are thereby required by the U.S. Clean Air Act to implement measures to reduce on-road emissions. The on-road transportation sector dominates fossil fuel-derived CO<sub>2</sub> emissions (ffCO<sub>2</sub>) in both cities, constituting 43% of LA's<sup>23</sup> and 38% of SLC's emissions.<sup>24</sup> On-road mobile sources also constitute the largest source of CO emissions in the counties where the two cities reside, comprising 47% and 56% of CO emissions in LA County and Salt Lake County, respectively.<sup>1</sup> The fleet composition of the two cities are different, which may affect their on-road combustion efficiency. SLC's fleet has a larger share of passenger trucks than cars (46% vs 41% respectively), while LA's fleet is dominated by passenger cars (36% trucks vs 57% cars) (Figure S1). Further, SLC's passenger cars are older on average than in LA, with mean ages of 9 and 7 years in SLC and LA, respectively (Figure S2).

California and Utah differ in the stringency of their statewide regulations concerning on-road emissions. The State of California was the first jurisdiction in the U.S. to regulate motor vehicle emissions of CO (1966) and CO<sub>2</sub>.<sup>25</sup> Since the 1960s, California has been granted waivers by the U.S. Environmental Protection Agency to write its own air pollutant regulations that are enacted separately from national laws and are more stringent. These policies include progressively stricter emission standards for vehicles sold in the state, on-board diagnostics (or "check engine" light) systems, and enhanced vehicle inspection/maintenance (I/M) programs intended to identify gross polluters (e.g., smog checks) in urban areas.<sup>26</sup> Further, the state also has incentive programs to encourage retirement of old vehicles and the purchase of low polluting vehicles.<sup>27</sup> In addition, California has strict mandates on fuel formulations, and introduced cleaner-burning, low sulfur fuel standards with its adoption of the LEV III standards in 2012. Thus, trends in LA's on-road CO/CO2 ratio should be decreasing in response to strict statewide emission regulations, incentivized fleet turnover and cleaner burning fuels.

In contrast, SLC is in the State of Utah, where vehicle sales and emission regulations generally follow federal policies that are less stringent and are usually adopted years after similar policies in California. Currently, Utah has ongoing incentive programs for repair or replacement of vehicles that fail I/M checks and offers incentives for the purchase of clean air vehicles,<sup>28</sup> albeit at a smaller scale than California. While smog checks are required in SLC as part of federally approved I/M programs since it is both an ozone and fine particulate matter nonattainment area, they are not a statewide requirement in Utah. Furthermore, federal Tier 3 fuel standards, which are highly similar to California's LEV III standard, were introduced in April of 2014.<sup>29</sup> As Utah is a small, somewhat isolated, market for vehicle fuel, the arrival of Tier 3 fuels occurred significantly later than the federal promulgation, with arrival of these fuels largely due to local political action and incentives. As a result, Tier 3 fuels were phased into the SLC market in 2020 and later, lagging California's adoption by over seven years. Hence, due to Utah's later and less stringent adoption of on-road emission regulations compared to California, we

expect that traffic combustion efficiency in SLC will not have improved as much as in LA.

2.2. On-Road Trace Gas Measurements. We measured on-road dry air mixing ratios of CO and CO<sub>2</sub> in LA and SLC using cavity ringdown spectrometers (Picarro, Sunnyvale, California) installed inside vehicle-based mobile laboratories following similar protocols as previous work in the two cities.<sup>30,31</sup> Measurements were made on weekdays between July 15-31, 2019 in LA and August 14-29, 2019 in SLC. We compared our observations to similar data collected between June 14 - July 7, 2013 in LA and August 9-17, 2013 in SLC.<sup>30,31</sup> Additionally, we collected similar measurements in LA on July 9-31, 2020 and July 15-16, 2021 to assess changes during the COVID-19 pandemic. In July 2020, the LA area was under stringent restrictions to prevent virus spreading, including closure of schools and indoor "non-essential" businesses, limited building capacities, quarantine requirements, travel restrictions, decreased public transit options, and state-wide recommendations to shelter in place.<sup>32,33</sup> By July 2021, state and local mandates had been lifted. Public transit use was 51% of July 2019 levels in July 2020, based on monthly bus and rail ridership on the LA Metro system.<sup>34</sup> By July 2021, public transit ridership had increased but had not recovered (64% of July 2019 levels)."

The mobile platform was a 2017 Mercedes Sprinter cargo van for the LA 2019–2021 surveys,<sup>35</sup> and a 2009 Hyundai Santa Fe Google Street View car for the SLC 2019 surveys.<sup>36</sup> The 2013 surveys utilized the same 2011 Ford Transit Connect van in both cities. The mobile laboratories were fabricated to stream ambient air into the analyzer with the inlet sampling air above the roof near the front of the vehicle. Geospatial coordinates and meteorological information were continuously recorded during the drives using compact GPS (Garmin GPS 16x in LA 2019–2021 and SLC 2013, Garmin 18x in SLC 2019) and weather sensors (LA only) mounted to the vehicle rooftops.

Traffic conditions were variable throughout the measurements, with a mix of congested and free-flowing traffic. Between 11 am-3 pm, we typically experienced stretches of free-flow until we reached congested areas (i.e., due to bottleneck areas or accidents). Congestion was less prevalent in 2020, as reflected by our overall faster speed. Our median speed was 5 mph faster in 2020 compared to 2019. Additionally, the data set captures some regular commuter rush hour traffic, which usually begins after 3 pm.

For year-to-year comparisons, data sets were filtered to only include overlapping locations (measurements within a 50 m buffer of each other). We also only compared data collected on freeways ("primary" roads as defined by the U.S. Census Bureau). After these filters, our data set represents approximately 470 km of road in LA and 60 km in SLC, most of which were sampled two or more times per survey. This includes segments of over ten freeways in LA (State Routes (SR-) 1, 22, 57, 60, 73, 91 and Interstate (I-) 5, 10, 105, 110, 210, 405 and 710) and two freeways in SLC (I-15 and 80). Our data set is representative of the mixture of emissions from vehicle tailpipes on the roadways which we measured, and we expect minimal influence from nonroad sources. One measurement day (August 16, 2013) in SLC was affected by smoke from a nearby wildfire and excluded from the analysis (Figure S3).

We followed instrument calibration and data processing protocols as described in previous studies where the 2013 data

sets were originally published.<sup>30,31</sup> The CO and CO<sub>2</sub> dry air mixing ratios were recurrently calibrated based on linear corrections between the Picarro G2401 measurements against standard tanks with known CO and CO<sub>2</sub> mixing ratios (Table S1). To synchronize the observations from the various instruments, data were integrated into 5-s averages and then gridded by averaging consecutive measurements into 100 m road intervals. Only daytime data collected between 11:00 and 16:00 (local time) was used in the analysis, when the planetary boundary layer is deep and air is well mixed. Nighttime and nonsummer measurements were excluded to avoid influence of biosphere respiration and shallow surface mixing layers, which would increase  $CO/CO_2$  levels and confound the signal from vehicle combustion efficiency. We calculated excess values above a background (denoted with subscript "xs"; e.g., CO<sub>2xs</sub>) for all gas mole fraction measurements by subtracting a regional background value from all observations. We estimate the background based on the "cleanest" measurements in our data set and by comparisons against CO and CO<sub>2</sub> observations upwind of the urban areas. Further details on our background characterization approach are described in the SI Text.

Given the diurnal and season variability in meteorology affecting  $CO/CO_2$  levels, we constrain the season and times of our analysis to summer daytime hours. Thus, we note that our study does not represent annually integrated CO/CO<sub>2</sub> signals, but instead year-to-year changes in the summer months only. Summertime is ideal for this analysis because of higher mixing layer heights and better mixing conditions, thereby reducing biases associated with pollutants getting trapped near the surface. Moreover, in the summer, we expect minimal influence of biospheric emissions in our on-road CO<sub>2</sub> signal since photosynthesis outpaces respiration.<sup>37</sup> Warmer temperatures also minimize the influence of cold engine start emissions, which would lead to lower combustion efficiency. For these reasons (mixing layer dynamics, biospheric fluxes, and cold engine starts), previous studies have observed higher  $CO/CO_2$ in the wintertime and other times of the day (mornings, evenings, and nighttime).<sup>30,31</sup> Thus, we limit our analysis to one season and only daytime hours to reduce the influence of these factors and focus on the effects of on-road vehicle combustion efficiency.

We expect our CO<sub>xs</sub> and CO<sub>2xs</sub> observations to represent only local emissions from vehicles on the road, with minimal influence from other emission sources. Biospheric  $CO_2$ emissions from respiration are outpaced by uptake from photosynthesis in the summer and, even in extreme scenarios, would be much smaller (less than 10 ppm) than the large  $CO_2$ signals we observe on the road (on the order of 100 ppm above background). Non-anthropogenic CO emissions (e.g., from the oxidation of biogenic VOC's) can be another source of CO in urban areas with high biogenic activity. However, it is not likely this has a notable impact on our on-road CO signal due to the localized, ground-level nature of our mobile measurements. This secondary CO is unlikely to stagnate on roadways, and would be dwarfed by the large signal from direct CO emissions from surrounding vehicles. Our measurements are collected directly at the surface, right at the source, making them highly localized and capturing the exhaust emissions in real time before they disperse and mix with other emissions.

#### 3. RESULTS AND DISCUSSION

3.1. Trends in  $CO_{xs}/CO_{2xs}$  between 2013 and 2019. Measurements of on-road  $CO_{xs}/CO_{2xs}$  ratios revealed con-





**Figure 1.** Histograms showing the distribution of  $CO_{xs}/CO_{2xs}$  for each mobile survey: (left panels) Los Angeles in (a) 2013 and (b) 2019, (right panels) Salt Lake City in (c) 2013 and (d) 2019. The solid vertical lines indicate the median and the dashed line indicates the mean value of each survey. For visualization purposes and comparison between surveys, the *x*-axis has been truncated. Inset maps on each panel show the full data distribution of each survey without truncation.



Figure 2. Maps showing the ratio of  $CO_{xs}/CO_{2xs}$  for each mobile survey: (left panels) Los Angeles in (a) 2013 and (b) 2019, (right panels) Salt Lake City in (c) 2013 and (d) 2019.

trasting temporal trends in the combustion efficiency of vehicle fleets in LA and SLC between 2013 and 2019 (Figure 1). Values for  $CO_{xs}/CO_{2xs}$  in LA and SLC were similar in 2013, but diverged in 2019. In LA, we observed a reduction in the on-road  $CO_{xs}/CO_{2xs}$  ratio from a median  $CO_{xs}/CO_{2xs}$  value of 5.0 ppbv/ppmv in 2013 to 2.6 ppbv/ppmv in 2019 (Figure 1a,b). In SLC, the median  $CO_{xs}/CO_{2xs}$  value increased from 4.1 ppbv/ppmv in 2013 to 6.4 ppbv/ppmv in 2019 (Figure 1c,d). The median  $CO_{xs}/CO_{2xs}$  ratio of each city was statistically different between years according to Mood's median test (*p*-value <0.05), indicating that the observed differences in the median values across years are unlikely to be due to random variation. Thus, traffic combustion efficiency had significantly improved in LA, but degraded in SLC between our measurement periods. Additionally, in LA, the  $CO_{xs}/CO_{2xs}$  ratios were more variable in 2013 compared to



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**Figure 3.** Histograms and maps of on-road  $CO_{xs}/CO_{2xs}$  observations in Los Angeles in July (a,b) 2019, (c,d) 2020, during COVID-19 pandemic related traffic reductions, and (e,f) 2021. The dashed lines on the histograms are the median  $CO_{xs}/CO_{2xs}$  values for each survey. The *x*-axes on the histograms have been truncated for visualization purposes.

2019, with the interquartile range decreasing from 3.3 in 2013 to 1.4 and 2019. The opposite was observed in SLC, where the interquartile range increased from 2.4 in 2013 to 5.8 in 2019. Furthermore, the data became less skewed in LA, with skewness values decreasing from 5.0 in 2013 to 2.2 in 2019, but in SLC, skewness values increased from 3.8 in 2013 to 6.9 in 2019. The changes in skewness are also exemplified by Lorenz curves and the Gini indices of each survey's  $CO_{2xs}$ , and  $CO_{xs}/CO_{2xs}$  values (Figure S4). Overall, the LA 2019 measurements had the lowest and least variable ratios and the SLC 2019 measurements had the highest and most variable ratios, with a maximum of 184 ppbv/ppmv. Taken together, our data shows that combustion efficiency increased from 2013 to 2019 in LA but decreased in SLC.

In LA, the reduction in  $CO_{xs}/CO_{2xs}$  ratio between the summers of 2013 and 2019 was generally observed across the entire basin, while the changes in SLC were more spatially heterogeneous (Figure 2). We observed several recurring CO<sub>xs</sub>/CO<sub>2xs</sub> hotspots in both 2013 and 2019, indicating persistent effects of traffic features such as steep roads and major freeway junctions on combustion efficiency. We considered hotspots as locations where CO<sub>xs</sub>/CO<sub>2xs</sub> exceeded the 95th percentile of observations in that city and year. In LA, CO<sub>xs</sub>/CO<sub>2xs</sub> ratios were elevated on steep roads, such as on SR-73 and SR-241, where CO<sub>xs</sub>/CO<sub>2xs</sub> exceeded 12 ppbv/ ppmv in 2019. This is likely because engine load is increased when driving upslope and thereby reduces the combustion efficiency. Additional reappearing hotspots were observed in LA near the junction of SR-57 and SR-60 near Pomona and near the SR-134 and SR-710 junction near Pasadena on both years. Combustion efficiency likely decreases near freeway

junctions since they often experience congestion, stop-and-go conditions and frequent acceleration as vehicles merge. Similarly, in SLC, hotspots were observed on the eastern portion of I-80 leading up to the Wasatch mountains. Additionally, new SLC hotspots were observed in 2019 that were not observed in 2013 that could potentially be due to construction or traffic conditions. Overall, SLC freeways had notably higher  $CO_{xs}/CO_{2xs}$  in 2019 compared to 2013, especially on I-15 and near its junction with SR-201 and I-80.

We compared the CO<sub>xs</sub>/CO<sub>2xs</sub> trends we observed to the California Air Resources Board's EMFAC model.<sup>38</sup> We downloaded annual CO and CO<sub>2</sub> emission estimates (in tons per year) for the South Coast Air Basin for the years 2013 and 2019-2021. We converted the emissions into molar units and calculated the CO/CO<sub>2</sub> ratio (in units of 1000 mol CO/ mol  $CO_2$ ) to match our observations in units of ppbv/ppmv. Based on the EMFAC output, the fleet-wide  $CO/CO_2$ decreased by 42% between 2013 and 2019. This corresponds well with our observed median change of 48% over the same time period. Thus, our observations and EMFAC agree that the fleetwide combustion efficiency improved between 2013 and 2019 in LA. Furthermore, assuming the reduction rate was linear and constant over the six years, this indicates a decreasing trend in  $CO/CO_2$  of -7.1% yr<sup>-1</sup> using EMFAC and -8.0% yr<sup>-1</sup> based on our observations. This is on par with earlier reports of a -7.8% yr<sup>-1</sup> trend from 1960 to 2010 based on regional atmospheric observations.<sup>9</sup> Based on EMFAC, annual on-road CO emissions decreased by 38% in 2019 relative to 2013 (or 28,500 tons CO yr<sup>-1</sup>) alongside these improvements in vehicle combustion efficiency in LA. Comparison of emission inventory estimates of SLC's on-



**Figure 4.** Box plots of hourly traffic speed, derived from the ratio of reported vehicle miles traveled (VMT) per vehicle hour traveled (VHT), for (a) Los Angeles freeways (I-5, I-405, I-605, I-210, SR-60, and SR-91) and (b) Salt Lake City freeways (I-15 and I-80) over the month each mobile survey was conducted. Only daytime hours are included (11 AM-4 PM local time). VMT and VHT data were downloaded from the Performance Measurement Systems data sources (CalTrans, 2022; UDoT, 2022).

road CO and  $CO_2$  (i.e., U.S. EPA's MOVES model) were not assessed in this study.

**3.2. Trends in Los Angeles CO\_{xs}/CO\_{2xs} during the COVID-19 Pandemic.** The COVID-19 pandemic allowed us to further study the effects of traffic conditions on vehicle combustion efficiency. Emergency physical distancing mandates were imposed in California starting in March of 2020 to reduce rates of virus transmission (CA Executive Order N-33-20). This resulted in an abrupt shift to remote work and learning, the closure of "non-essential" businesses and entertainment venues, and strict limitations on domestic and international travel. Consequently, there was a drastic reduction in freeway traffic and congestion in the year 2020.

We repeated the LA on-road measurements in July 2020 and 2021 to assess how the combustion efficiency was affected by the sudden changes in commuter traffic (Figure 3). We found that the median CO<sub>xs</sub>/CO<sub>2xs</sub> increased from 2.7 ppbv/ppmv in 2019 to 6.1 ppbv/ppmv in July 2020, indicating a downturn in the fleet combustion efficiency during COVID-19 restrictions. The  $CO_{xs}/CO_{2xs}$  observations were also more variable in 2020, with the interquartile range increasing from 1.4 in 2019 to 3.7 in 2020. As pandemic-related mobility restrictions were gradually relaxed, traffic patterns eventually returned to prepandemic levels. By July 2021, on-road CO<sub>xs</sub>/CO<sub>2xs</sub> decreased to a median value of 2.5 ppbv/ppmv and an interquartile range of 1.4 which coincide with the prepandemic (2019) observations. This indicates that the increased  $CO_{xs}$ / CO<sub>2xs</sub> ratios in 2020 were temporary and returned to the previous state of combustion efficiency by 2021.

The stark reductions in LA's traffic combustion efficiency in 2020 indicate that on-road  $CO_2$  and CO emissions were substantially affected by changing driving patterns during the COVID-19 pandemic. In July 2020, LA's on-road  $CO_{2xs}$  levels were reduced by 60% relative to July 2019, with a near complete rebound by July 2021 (Figure S5). We attribute the 2020  $CO_{2xs}$  reductions to the decrease in the number of vehicles on the road, since  $CO_2$  emissions are directly proportional to the amount of fuel burned. However, on-road  $CO_{xs}$  levels did not show a significant change between 2019 and 2021 (Figure S5). This implies that worsened combustion efficiency maintained the typical on-road CO levels despite there being fewer vehicles on the road in 2020.

Using the median  $CO_{xs}/CO_{2xs}$  observations in 2019 and 2020 as emission factors and on-road  $CO_2$  emissions from

EMFAC (65.7 million tons in 2019 and 58.5 million tons in 2020), we calculate that the worsened combustion efficiency led to 130,000 more tons of CO emitted in 2020 than would have been emitted if the combustion efficiency had remained at the 2019 level. This amounts to 20% of the South Coast Air Basin's total annual CO emissions (653,000 tons  $yr^{-1}$ ), based on 2017 CO inventory estimates.<sup>39</sup> Thus, the less efficient vehicle combustion during the pandemic led to a marked effect on CO emissions relative to the total CO budget. The potential causes of the decreased combustion efficiency in 2020 are discussed in the following section.

3.3. Evaluation of the Potential Contributors to CO<sub>xs</sub>/ CO<sub>2xs</sub> Trends. In summary, our measurements indicate an improvement in traffic combustion efficiency in LA since 2013, except for during the COVID-19 pandemic when combustion efficiency worsened. Conversely, combustion efficiency in SLC showed a decline between 2013 to 2019. Policy interventions and technological advances should be reducing CO<sub>xs</sub>/CO<sub>2xs</sub> in both cities, with potentially stronger reductions in LA than SLC due to stricter and earlier adoption of regulatory measures in California. However, our measurements indicate a more complex interplay of factors because  $CO_{xs}/CO_{2xs}$  increased in SLC and during COVID-19 in LA, opposing the expected decrease with emissions control measures. In this section, we discuss traffic and fleet characteristics that increase  $CO_{ys}/CO_{2ys}$ and evaluate their potential contributions based on our observations, relevant literature and the vehicle composition in LA and SLC.

An increase in heavy-duty vehicle activity would decrease  $CO_{xs}/CO_{2xs}$  because diesel engines produce substantially less CO per unit of fuel burned than gasoline-powered engines.<sup>40–43</sup> Thus, heavy-duty vehicles cannot explain the  $CO_{xs}/CO_{2xs}$  increase we observed during COVID-19 and in SLC unless real-world  $CO/CO_2$  emission ratios from heavy-duty vehicles differ grossly from expectation. Additionally, cold engine starts lead to higher  $CO_{xs}/CO_{2xs}$  but are unlikely to occur during summer and on interstate freeways where our measurements took place. Construction activity may cause higher  $CO_{xs}/CO_{2xs}$  due to less efficient off-road equipment, but would have episodic effects on the data, not an overarching shift in the distribution as we observed.

3.3.1. Vehicle Speeds. In general, CO and CO<sub>2</sub> emission rates increase at low (<30 mph) and high (>55 mph) speeds.  $^{40,44-46}$  However, the effect of speed on CO/CO<sub>2</sub> ratios

varies with vehicle class, fuel type, and age (Figures S6, S7). Vehicles were driving faster during the two surveys in which we observed increases in  $CO_{xs}/CO_{2xs}$  (LA 2020 and SLC 2019). Based on traffic count data<sup>47,48</sup> the median speed increased significantly in LA by 5 mph in 2020 compared to 2019 (Figure 4; p-value <0.05 using Mood's median test). Other reports also indicated faster driving speeds and more aggressive driving in 2020, which led to higher rates of severe crashes despite fewer vehicles on the road.<sup>49,50</sup> When traffic conditions returned to prepandemic levels in 2021, both the median traffic speed and the CO<sub>xs</sub>/CO<sub>2xs</sub> values returned to 2019 levels. In SLC, median traffic speeds increased by 3 mph in 2019 compared to 2013, coinciding with increased CO<sub>xs</sub>/CO<sub>2xs</sub> ratios. This may be due to a speed limit increase on I-15 which was implemented in 2015.51,52 While we did not measure  $\text{CO}_{xs}/\text{CO}_{2xs}$  in SLC in 2020–2021, it is notable that the increasing speed trend continued during those years.

We tested the effect of faster speeds on  $CO/CO_2$  using the California Air Resources Board's EMFAC model by simulating annual CO and CO<sub>2</sub> emissions if all vehicles were driving in their nighttime speed conditions, which we assume to represent free-flow traffic. Based on this scenario analysis, EMFAC predicts that  $CO/CO_2$  emission ratios decrease by 12% at faster speeds (Table S3; Figure S6). However, EMFAC only models emissions for speeds up to 70 mph and many drivers commonly exceed this speed. Some studies using portable emissions measurement systems installed on running vehicles have found that CO/CO<sub>2</sub> ratios increase at faster speeds, especially when the driving style is aggressive, defined by faster speeds and bursts of acceleration. 53-56 However, other studies (and EMFAC2021) have observed that CO/CO<sub>2</sub> decreases or does not change significantly with higher speeds.,44,46,57 The discourse is probably because the effect of faster speeds on CO/CO<sub>2</sub> emissions varies by vehicle fuel, class, age, and driving style. Based on EMFAC2021, CO/CO<sub>2</sub> decreases with speed for modern passenger cars, but increases for some light-duty trucks and older vehicles (Figures S6 and S7). SLC's fleet mix has more passenger trucks than LA (Figure S1) and is older (Figure S2). Thus, speed could be an important factor affecting our measurements, since  $\text{CO}_{xs}/$ CO<sub>2xs</sub> increased for both surveys in which speed increased (LA 2020 and SLC 2019). However, the impact of speed is dependent on the fleet composition (i.e., vehicle age, fuel, and category) and likely is not the only factor contributing to CO<sub>xs</sub>/CO<sub>2xs</sub> increases.

3.3.2. Vehicle Age. Older vehicles on the road would lead to increases in CO<sub>xs</sub>/CO<sub>2xs</sub> ratios (Table S3), especially if they are driving fast (Figure S7). Newer vehicle models are continually improved to emit less pollutants, while older models may have outdated emission control technology.<sup>58</sup> For instance, California's Low Emission Vehicle (LEV) standards have mandated increasingly lower CO emission rates across model year generations: pre-1993 (pre-LEV standards), 1994-2003 (LEV I), 2004-2014 (LEV II), and 2015-2025 (LEV III). Further, as vehicles age and/or accumulate mileage, the effectiveness of their on-board emission control technology deteriorates.<sup>58,59</sup> It is possible that older vehicles played a role in the two surveys in which  $CO_{xs}/CO_{2xs}$  increased. On average, SLC has older vehicles than LA (Figure S2). During the COVID-19 pandemic, older vehicles may have become more prominent on the road, which would contribute to the increased  $CO_{xs}/CO_{2xs}$  ratios we observed. While many Californians switched to teleworking during the pandemic,

"essential workers" resumed work that was essential to maintain safety, health, and sanitary services.<sup>60</sup> Those driving in 2020 may have largely been service and/or blue-collar workers who may have limited ability to purchase newer vehicles. Further, public transportation use plummeted during the pandemic due to physical distancing recommendations. Regular transit users may have increased their driving of old vehicles if they could not afford a more modern car. We simulated the impact of older vehicles in EMFAC and found that if the 2020 traffic fleet reverted to the 2013 fleet (an oldervehicle scenario), on-road CO/CO<sub>2</sub> ratios would increase by 84% relative to the default scenario (Table S3). If the older vehicles were driving fast and/or with an aggressive driving style, these effects occurring simultaneously could compound  $CO_{xs}/CO_{2xs}$  increases (Figure S7).

The combination of an older vehicle fleet mix and higher speeds in SLC relative to LA, and during 2020 in LA relative to 2019 and 2021, is the most likely explanation for our realworld  $CO/CO_2$  observations (Table S3). While we recognize that for most of the vehicle fleet, speed has a decreasing effect on  $CO/CO_2$  in theory, the characteristics of the SLC fleet (older and more light duty trucks) and the circumstances around the COVID-19 pandemic could lend toward increased  $CO/CO_2$  at faster speeds, especially if the driving style is aggressive. Our data set captures the net fleet-wide  $CO/CO_2$ , and thus does not distinguish which vehicles caused the increase. Further work using portable emissions measurement systems or chassis dynamometer tests is needed to isolate the vehicle-specific effects. Such tests were used to inform emission rates in EMFAC but are limited for this comparison because (1) emissions under real-world driving conditions differ from the controlled test cycles on which EMFAC is based (i.e., higher than 70 mph speeds and more aggressive driving styles) and/or (2) because the pandemic led to broad fleet composition changes that were not captured by the model. Further, very few studies measuring emissions under real-world driving directly report CO/CO2 ratios.53 Nonetheless, our observations suggest that the complex effects of speed on emissions negatively impact combustion efficiency. Further research is needed, especially on emissions and combustion efficiency at high vehicle speed (>70 mph).

**3.4. Implications for Fossil Fuel CO<sub>2</sub> Quantification.** Our observations imply that the robustness of the CO/CO<sub>2</sub> ratio as a tracer of fossil fuel-sourced CO<sub>2</sub> (ffCO<sub>2</sub>) is diminishing as vehicle CO emissions are decreasing. Reliable ffCO<sub>2</sub> tracers are critical for studies that attempt to quantify anthropogenic CO<sub>2</sub> emissions mostly coming from fossil fuels, given that the biosphere contributes significantly to CO<sub>2</sub> emissions, even in cities.<sup>37</sup> Since incomplete combustion of fossil fuels results in coemitted CO and CO<sub>2</sub>, the ratio of the two gases has been used to distinguish ffCO<sub>2</sub> emissions from non-anthropogenic CO<sub>2</sub> sources.<sup>17,18,61-63</sup> The ffCO<sub>2</sub> enhancement is calculated as

$$ffCO_2 = \frac{CO_{obs} - CO_{bg}}{R_{CO/ffCO_2}}$$
(1)

where  $CO_{obs}$  is the measured CO mixing ratio,  $CO_{bg}$  is the CO background (usually determined from a remote or upwind site), and  $R_{CO/ffCO_2}$  is the ratio between CO and ffCO<sub>2</sub> in units of ppbv/ppmv. Ideally,  $R_{CO/ffCO_2}$  would be calculated based on the correlation between CO and the radiocarbon-based estimate of the ffCO<sub>2</sub> signal, since the radiocarbon

isotope is the most direct atmospheric proxy for fossil fuel emissions.<sup>17</sup>

The uncertainty in ffCO<sub>2</sub> calculations using this approach depends on the variability in  $R_{CO/ffCO_2}$  for the particular location and time period of the study. For instance, in previous work, an  $R_{CO/ffCO_2}$  value of 14 ± 2 ppbv/ppmv resulted in ffCO<sub>2</sub> values that varied by approximately  $\pm 15\%$  based on the upper and lower  $R_{CO/ffCO_2}$  values and background conditions in Sacramento, CA in 2009.<sup>17</sup> However, with CO emissions declining in urban areas, uncertainty in ffCO<sub>2</sub> would increase as  $R_{\rm CO/ffCO_2}$  approaches zero. Using the variability in measured on-road  $\mathrm{CO}_{xs}/\mathrm{CO}_{2xs}$  ratios to quantify uncertainty in  $R_{CO/ffCO_2}$  (i.e., 3.0 ± 1.6 in LA 2019), the resulting ffCO<sub>2</sub> values vary by 34–114%. This large uncertainty is conservative, given that LA 2019 was our least variable survey and other years and SLC had larger standard deviations ranging from 4.7 to 17.1 ppbv/ppmv. Given the spatial and temporal variability in the ratio observed in this study, the uncertainty in ffCO<sub>2</sub> using this method is substantial. This approach requires a robust correlation between CO and ffCO2, which we did not observe on the road (Figure S8). In our observations, we expect that all of the CO<sub>2xs</sub> we measured on-road was from fossil sources, yet CO<sub>xs</sub> and CO<sub>2xs</sub> were not strongly correlated, with  $R^2$  values less than 0.19 (Figure S8). The correlation weakened over time. The 2019-2021 observations had lower  $R^2$  values (ranging from 0 to 0.16) than the 2013 measurements in both cities (R ranging from 0.17 to 0.19). These weak correlations underscore the diverging trends in on-road emissions, where CO<sub>2</sub> is always being produced while fuel is being burned, but CO should be destroyed by the catalytic converter and is only emitted sporadically when suboptimal conditions are present. This is further evidenced by the skewed distribution of our CO measurements (Figure S5). Thus, for vehicles with internal combustion engines, weakening correlations between on-road CO and CO<sub>2</sub> levels are expected with effective CO control efforts and as technology advances over time. Our findings are in line with previous studies that described inconsistencies and large uncertainties in CO/CO<sub>2</sub> that make CO an unreliable tracer for ffCO<sub>2</sub> emissions on its own.<sup>44,64,65</sup> Other trace gas species such as  $NO_2$  may be suitable alternative tracers,<sup>66,67</sup> but updated studies evaluating such proxies against radiocarbon observations, the most direct tracer for  $ffCO_{2i}$  are urgently needed to ensure robust tracking of climate change mitigation measures.

In summary, using on road measurements of CO and CO2 mole fractions, we observed changes in vehicle combustion efficiencies in two western U.S. cities (Los Angeles, CA and Salt Lake City, UT) over 2013 and 2021, a period that includes substantial changes in vehicle age and speed. Our measurements show that stricter emission regulations and mitigation incentives successfully lowered on-road CO emissions across summers over a six-year period in LA. In contrast, the combined effects of traffic conditions and the fleet composition led to a net worsening of fleet combustion efficiency in SLC, and during the COVID-19 pandemic in LA. These on-road mobile measurements capture the complex mix of sources and drivers in the real world that may differ from the model-based predictions. Future work should further evaluate the effects of traffic conditions on urban emissions and policymakers should consider the negative effects of elevated driving speeds on air quality. Furthermore, the success of CO emission regulations

will make it more challenging for studies to apply CO as a tracer for quantifying fossil fuel  $CO_2$  emissions from cities.

#### ASSOCIATED CONTENT

#### Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.4c11701.

Vehicle fleet information for each urban area, further details about measurement and data procedures, EMFAC emission rates analysis,  $CO:CO_2$  correlation plots (PDF)

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#### Notes

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