Environmental Science & Technology

pubs.acs.org/est

Article

Composition, Emissions, and Air Quality Impacts of Hazardous Air Pollutants in Unburned Natural Gas from Residential Stoves in California

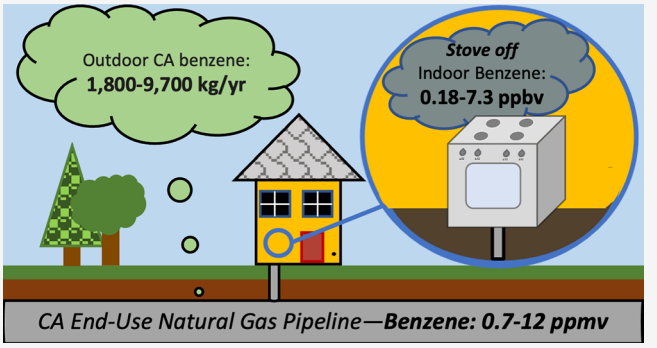
Eric D. Lebel,* Drew R. Michanowicz, Kelsey R. Bilsback, Lee Ann L. Hill, Jackson S. W. Goldman, Jeremy K. Domen, Jessie M. Jaeger, Angélica Ruiz, and Seth B. C. Shonkoff

Cite This: *Environ. Sci. Technol.* 2022, 56, 15828−15838

Read Online

ACCESS | Metrics & More | Article Recommendations | Supporting Information

ABSTRACT: The presence of hazardous air pollutants (HAPs) entrained in end-use natural gas (NG) is an understudied source of human health risks. We performed trace gas analyses on 185 unburned NG samples collected from 159 unique residential NG stoves across seven geographic regions in California. Our analyses commonly detected 12 HAPs with significant variability across region and gas utility. Mean regional benzene, toluene, ethylbenzene, and total xylenes (BTEX) concentrations in end-use NG ranged from 1.6−25 ppmv—benzene alone was detected in 99% of samples, and mean concentrations ranged from 0.7−12 ppmv (max: 66 ppmv). By applying previously reported NG and methane emission rates throughout California's transmission, storage, and distribution systems, we estimated statewide benzene emissions of 4,200 (95% CI: 1,800−9,700) kg yr^{-1} that are currently not included in any statewide inventories—equal to the annual benzene emissions from nearly 60,000 light-duty gasoline vehicles. Additionally, we found that NG leakage from stoves and ovens while not in use can result in indoor benzene concentrations that can exceed the California Office of Environmental Health Hazard Assessment 8-h Reference Exposure Level of 0.94 ppbv—benzene concentrations comparable to environmental tobacco smoke. This study supports the need to further improve our understanding of leaked downstream NG as a source of health risk.



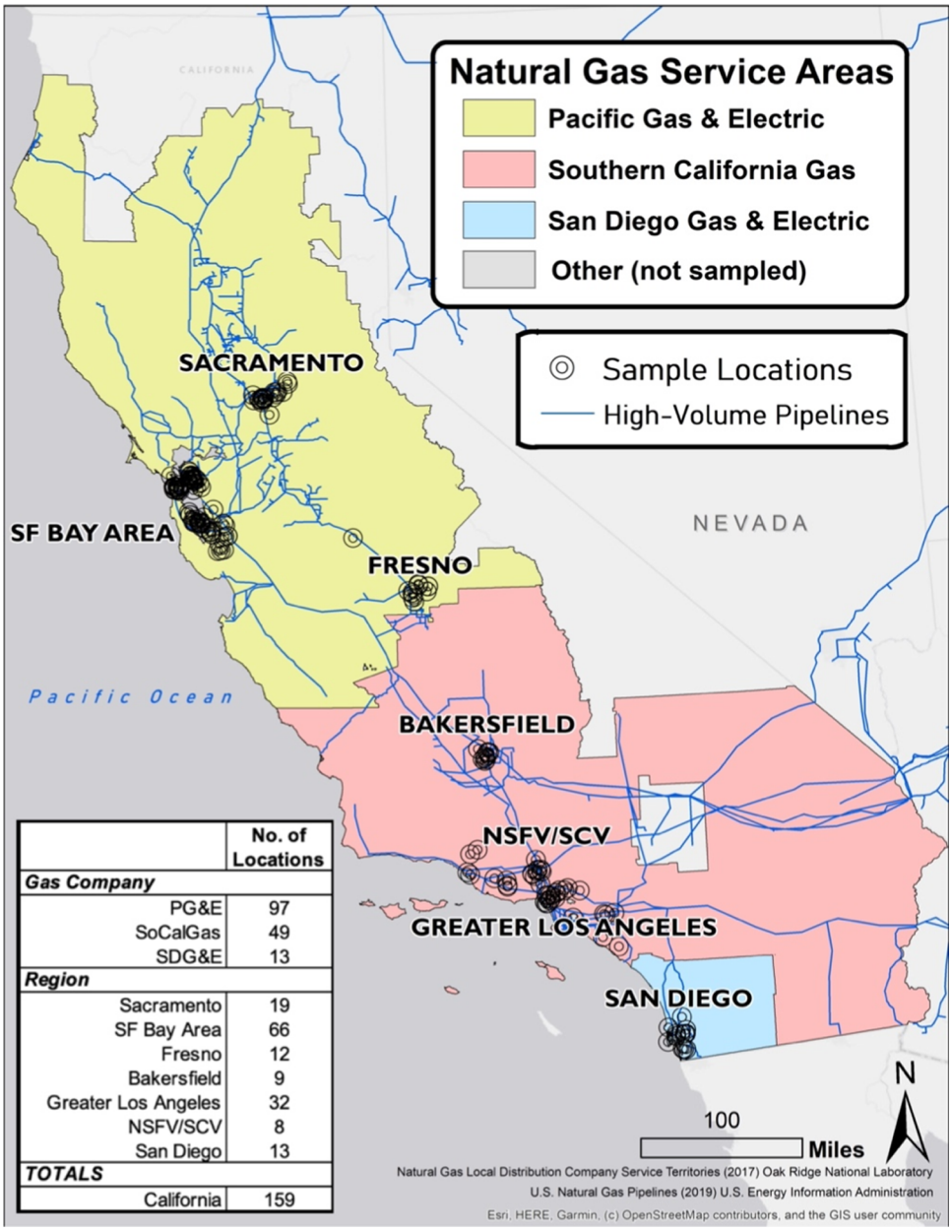
KEYWORDS: *BTEX, benzene, downstream, fossil fuels, natural gas leak, cooking, hazardous air pollutants, indoor air quality, regional BTEX inventories*

INTRODUCTION

In 2021, the United States consumed approximately 30.2 trillion cubic feet of processed natural gas (NG), comprising 32% of total U.S. energy consumption, of which 15% was consumed by an estimated 74.6 million residential households.[1−3] Non-methane volatile organic compounds (NMVOCs) are naturally present in unprocessed NG;[4−9] however, very little data exists on the chemical composition and concentrations of hazardous air pollutants (HAPs) in processed NG at the point of end use.[10] Moreover, the indoor air quality impacts from fugitive NMVOC or HAP emissions associated with known NG leakage or incomplete combustion pathways are understudied, in part, due to a lack of chemical characterization of downstream NG.[11]

While NG transmission pipelines have some NMVOC restrictions to prevent operational failures (see Federal Register Vol 71 No 120), there are no corresponding limits on HAPs in transmission pipeline gas in the United States.[12−16] Many NMVOCs are hazardous to human health and therefore have been deemed HAPs under the U.S. Clean Air Act, which is further exemplified by various state, federal, and international health-based guidelines for exposure to these compounds.[17,18] For example, NMVOCs contribute to the formation of ozone, a constituent of photochemical smog, and fine particulate matter ($PM_{2.5}$).[19−21] Benzene is a known human carcinogen that increases the risks of developing leukemia.[22] One study detected benzene, toluene, ethylbenzene, and xylenes (BTEX) and other health-damaging NMVOCs in downstream NG;[23] however, these data were not quantitative and only included two samples. Recently, Michanowicz et al.[24] found extensive NMVOCs in distribution NG in Massachusetts; however, measurements were confined to a region that is geographically far from hydrocarbon

Received: April 13, 2022
Revised: September 30, 2022
Accepted: September 30, 2022
Published: October 20, 2022



	No. of Locations
Gas Company	
PG&E	97
SoCalGas	49
SDG&E	13
Region	
Sacramento	19
SF Bay Area	66
Fresno	12
Bakersfield	9
Greater Los Angeles	32
NSFV/SCV	8
San Diego	13
TOTALS	
California	159

Figure 1. Spatial distribution of the 159 households sampled in California. The points show the location of each of the samples. The shading shows gas utility service territories in each region, and the blue lines represent high-volume pipelines traversing California and neighboring states. Measurements were taken in the following gas companies' territories: Pacific Gas and Electric (PG&E)—yellow; Southern California Gas Company (SoCalGas)—red; and San Diego Gas & Electric (SDG&E)—blue. Samples were grouped into one of 7 different regions which we used throughout this study: San Francisco (SF) Bay Area, Sacramento, Fresno, Bakersfield, North San Fernando/Santa Clarita Valleys (NSFV/SCV), Greater Los Angeles (excluding NSFV/SCV), and San Diego.

production or underground NG storage in depleted oil reservoirs—two features present throughout California that may impact NMVOC concentrations in NG. There are also limited publicly available NMVOC composition data for the transmission and storage sector,[25] and it is unclear the degree to which these trace gases persist in the end-use gas distribution system.

Residential NG saturation in California is the second highest in the United States, with 88% of all households (11.5 million in total) having NG service in 2020. Typically, 70% of these homes cook with an NG-powered stove or oven, representing the highest percentage of gas stove users in the country.[1] Driven in part by concerns regarding the climate-forcing nature of methane, multiple studies in California have quantified NG emissions from buildings and appliances. Fischer et al.[26] measured NG appliance leakage in 75 homes in California and found that chronic, low-level leaks can persist undetected directly in residential homes and that 15% of total NG methane emissions in California could be attributed to fugitive post-meter (i.e., in-home) emissions. Lebel et al.[27,28] observed that at least 75% of total emissions from gas stoves and storage water heaters originate while they are off.[27,28] These leakage profiles, in combination with uncertainties related to the effectiveness of NG odorants as exposure deterrents,[24,29−34] support the need for further study of potential risks of exposure

https://doi.org/10.1021/acs.est.2c02581
Environ. Sci. Technol. 2022, 56, 15828−15838

to health-damaging compounds contained in end-use NG in California.

In this study, we adopted canister collection methods with gas chromatography−mass spectrometry (GC−MS) NMVOC analysis from Michanowicz et al.[24] to determine the chemical composition of unburned NG from downstream NG supply in seven distinct geographic locations in the state of California.[24] Our goals for studying the gas stream at the point of end use were twofold: (1) To understand the contribution of NMVOCs to urban air quality associated with leakage throughout the downstream NG supply chain, and (2) To understand potential indoor exposures from known leakage pathways. To begin to address goal 1, we combined these NG composition data with existing California-specific NG leakage data to estimate unaccounted-for BTEX emissions for the entire state and for certain individual source types within the downstream infrastructure. To address goal 2, we estimated indoor residential ambient benzene concentrations attributable to published rates of NG leakage from household gas stoves and ovens while off across various scenarios.[35,36] We compared these indoor air concentrations of benzene to the California Environmental Protection Agency Office of Environmental Health Hazard Assessment (OEHHA) benzene Reference Exposure Level (REL) to evaluate health risks from this understudied environmental exposure.[37]

METHODS

Data Collection. We collected 185 unburned NG samples from 159 unique residential NG stoves in California. We used passivated 1 L SUMMA or Silonite-lined Entech canisters with Teflon tubing to form a direct in-line connection with a single-stove burner NG outlet orifice. NG samples were analyzed by two labs based in California with Environmental Laboratory Accreditation Program (ELAP) certification. We used two labs to accommodate canister availability (Lab 1: n = 54; Lab 2: n = 131).

NMVOCs for all samples were analyzed using U.S. Environmental Protection Agency Method TO-15, which calls for sample separation and analysis by gas chromatography and mass spectroscopy.[38] Samples were analyzed by the lab within 30 days of collection—the maximum hold time specified by the TO-15 method during which concentrations are considered stable. The TO-15 analyte list included 69 compounds from Lab 1 and 63 compounds from Lab 2. We quantified a total of 76 unique compounds using TO-15 across the two laboratories, including 56 compounds analyzed by both labs (see Table S1 for a complete list). Each sample was also tested using Method ASTM D1946 to quantify methane and ethane content in unburned NG as well as elemental nitrogen and oxygen for all but 27 samples.[39] These data were used for quality control to ensure pure NG sample collection and to verify minimal ambient air intrusion during sampling.

We collected samples using a similar methodology as described in detail in Michanowicz et al.[24] and briefly summarized here. First, we removed gas stove flame spreader plates and metal grates to expose the NG outlet orifice. We then applied Teflon-lined tubing of 1/4" inner diameter to tightly enclose the NG outlet orifice, bypassing any ignition sources and minimizing the potential for NG leakage or ambient air entrainment. We then turned the gas flow knob between medium and high for the entirety of the sampling event. After flushing the hosing, we immediately connected it to the sampling canister provided by the lab via a cone washer and screw nut or quick release connection. We filled canisters until the vacuum on the canister was between −5 and 0 in Hg, taking approximately 20−30 s. For safety, we deployed a Bascom Turner Gas Rover or similar device near the stove to ensure the concentration of leaked NG did not approach the lower explosive limit of methane (5% v/v gas), and we turned on range hoods when present during and after the sampling process to reduce indoor air concentrations of NG.

We intentionally designed this measurement campaign to sample NG in major urban areas serviced by the three main NG utilities in California: Pacific Gas and Electric (PG&E), Southern California Gas Company (SoCalGas), and San Diego Gas & Electric (SDG&E). In all, we collected samples from 16 counties across California from these three utilities and grouped samples into seven regions we defined: the San Francisco Bay Area (San Francisco, San Jose, Oakland, and nearby cities); Sacramento and Fresno [PG&E]; Greater Los Angeles; the North San Fernando and Santa Clarita Valleys; and Bakersfield [SoCalGas]; and San Diego [SDG&E]. We collected samples between February and September 2021 (Figures 1 and S2).

Quality Control and Data Analysis. Data from our 185 samples are reported as pure gas mole fractions (concentrations) in parts per billion by volume (ppbv). Lab 1 reported directly in ppbv, while Lab 2 required conversion from mass concentrations to volumetric concentrations (ppbv) using the molecular weight of the compound and assuming normal temperature (298 K) and pressure (101.3 kPa). For values below the lab's internal limit of detection, we took a conservative approach and assigned these data as zero.[40]

We took several quality control measurements throughout the study, in addition to blanks and other quality checks performed by the labs. First, to compare our NG samples to background air collected by the same method, we collected one field blank by attaching the hosing to the stove and not turning the gas on and one ambient air blank by opening the can in the kitchen space with no hosing attached, both of which did not detect target analytes. Second, we used both methane and elemental nitrogen (N_2) as source signature tracers—methane as proxy for NG, and nitrogen as proxy for ambient air. Using these fixed gas measurements, we conducted a k-means cluster analysis to exclude samples that entrained a substantial amount of ambient air. Based on our analysis, we identified two k-clusters and excluded the cluster with the lowest methane and highest nitrogen values (Figure S1). To control for samples that did not have a paired nitrogen, nine samples were additionally excluded that had a methane measurement within the range of the excluded k-cluster. The mean of benzene and total BTEX of the excluded samples were less than the mean of the included samples; this is unsurprising, given the assumed dilution effect from ambient air intrusion (Figure S1).

We also collected a series of samples to quantify variability across our study. These included duplicate samples (collected consecutively at a single location) analyzed by different labs; duplicate samples analyzed by the same lab; and repeated samples that were collected in the same households several months apart (Note S1). In our analysis, duplicates collected on the same day were treated as replicate samples; therefore, we selected only the first of the two samples to be used in our analyses. For the times we collected repeat samples in different months, we treated them as independent samples for regional

https://doi.org/10.1021/acs.est.2c02581
Environ. Sci. Technol. 2022, 56, 15828−15838

and statewide means as household-level factors should not influence gas composition.

Regional and Statewide Emissions Estimates. To evaluate the contribution of downstream NG emissions to regional and statewide benzene and BTEX budgets, we used methane emission estimates from previous studies of the downstream NG sector in California to scale bootstrapped regional and statewide mean concentrations of benzene and total BTEX measured in our study.[26,28,41−44] Emissions rates of benzene and total BTEX were calculated using the following equation adapted from Marrero et al.[6]

$$E_{\mathrm{NMVOC}} = E_{\mathrm{methane}} \times C_{\mathrm{NMVOC}} \left(\frac{MW_{\mathrm{NMVOC}}}{MW_{\mathrm{methane}}} \right) \quad (1)$$

where E_{NMVOC} represents the NMVOC mass flow rate (kg yr^{-1}); E_{methane} represents the methane mass flow rate (Gg yr^{-1}) derived from existing literature;[26,28,41−44] C_{NMVOC} represents the state or regional concentration of NMVOCs in the pipeline NG (ppmv); and MW_{NMVOC} and MW_{methane} represent the molecular weights of the NMVOC and methane, respectively (g mol^{-1}). This relationship assumes that the concentration of methane in pipeline NG is 100%, and as such, we are slightly underestimating total NMVOC emissions. Scaled benzene emissions (E_{NMVOC}) assume a benzene-to-methane ratio that is conserved throughout the distribution pipeline network and likely underestimates NMVOC emissions using methane flux rather than NG flux. The emissions estimates also assume no differential mechanisms of transport and fate between methane and benzene emissions (e.g., soil adsorption)—a limitation of this methodology.

Regional and distribution pipeline methane emissions were taken from available data sources in the literature to scale our NMVOC emissions. NMVOC emissions for the North San Fernando and Santa Clarita Valleys specifically were estimated using distribution pipeline NG emissions from SoCalGas for zip codes in this region (Figure S4) and NMVOC measurements from our samples in the North San Fernando and Santa Clarita Valleys.

Indoor Air Quality Modeling. To better understand the potential indoor air quality implications of benzene content in end-use NG leaked from gas stoves while turned off, we modeled ambient benzene concentrations in the kitchen and compared the results to California's Reference Exposure Levels (REL). We modeled 140 scenarios that varied unburned NG leakage rates (median: 24 mg hr^{-1} and 95th percentile: 282 mg hr^{-1}, from Lebel et al.[28]), benzene concentrations in unburned gas (median and 95th percentiles calculated from this study), and household parameters (Table S2). We used CONTAM—a multizone, whole-building model developed by the National Institute of Standards and Technology—that can be used to estimate contaminant concentrations in the indoor environment using information about the structure, ventilation, airflow, and contaminants.[35,36,45]

Here, as an introductory assessment of the impact of leaked NG on indoor air quality, we conservatively focused only on leakage from a single gas stove/oven while off (steady-state off) and for only one pollutant: benzene. This modeling approach almost certainly underestimates the true quantity of behind-the-meter NG emissions which also may include leaks from other gas appliances and emissions of natural gas when the stove/oven is on.

We calculated benzene steady-state-off leakage rates from stoves using the following relationship

$$V_{\mathrm{benzene}} = V_{\mathrm{methane}} \times C_{\mathrm{benzene}} \quad (2)$$

where V_{benzene} represents the volumetric emissions of benzene from the leaking stove (nL hr^{-1}); V_{methane} represents the volumetric methane emissions rate from the leaking stove (mL hr^{-1}); and C_{benzene} represents the concentration of benzene in the the pipeline gas (ppmv), again assuming that the concentration of methane in pipeline NG is 100%. This enables us to directly convert the volumetric flow of methane, V_{methane}, to a volumetric flow of benzene, V_{benzene}, at a given concentration of benzene. Data on methane emission rates from leaking stoves were obtained from Lebel et al.,[28] who measured methane emission rates from 53 gas stoves in California using a chamber-based approach. Benzene concentrations are from this study. This relationship assumes that the emission rate of methane is equivalent to the emission rate of NG. This is a conservative assumption as distribution system NG was measured to be 90−95% methane in this study (Figure S5).

We modeled a range of household parameters and emissions rates; a complete list is in Table S2. We chose a range of households that reflects California's housing stock from the 2019 Residential Appliance Saturation Survey.[46] This included a mobile/manufactured home, an attached home, a detached single-family home, and a six-unit apartment building. Building designs were selected from the "Suite of Homes Representing the U.S. Housing Stock."[47,48] Within each household type, we varied the air change rate, the methane emission rate, and the benzene concentration in the pipeline gas. For comparison's sake, we used only two air change rates—the buildings' natural ventilation air change rate (i.e., infiltration only) and the minimum recommended air change rate set by the American Society of Heating, Refrigerating and Air-Conditioning Engineers (ASHRAE 62.2). The ASHRAE standard was achieved by parameterizing HVAC system settings to input the appropriate volume of air changes per hour. The methane emission rate and benzene concentration rate were used together to calculate the benzene emissions rate via eq 2 (Table S2). All simulations were run under steady-state airflow conditions at normal temperature (298 K) and pressure (101.3 kPa).

Across our simulations, we assumed that benzene was not reactive because the atmospheric lifetime of benzene is several days and the indoor concentrations of the hydroxyl radical ($^{\bullet}OH$) are similar to outdoors.[49,50] Following previous studies, we also assumed that surface deposition would not substantially impact indoor concentrations.[51,52] We assumed that there was no demand-controlled mechanical kitchen hood active during simulations, since our source was only fugitive emissions while off.

RESULTS AND DISCUSSION

Overview. We collected 185 individual samples of unburned NG from 159 unique residential stoves across seven distinct geographic locations spanning 16 counties throughout California (Figure 1). After eliminating blanks, duplicate samples, and low-methane samples that did not meet QC standards (see Materials and Methods), we retained 160 samples for analysis. Of the 76 NMVOCs analyzed, 21 unique constituents were detected in end-use NG in California (see Table S1 for a full list of pollutants). Of these, 12 carried a

https://doi.org/10.1021/acs.est.2c02581
Environ. Sci. Technol. 2022, 56, 15828−15838

HAP designation and therefore are known to cause short-term and chronic adverse health impacts.[53] Six NMVOCs were detected in more than 98% of NG samples, four of which are HAPs: benzene, toluene, m,p-xylene, and hexane (Figure 2).

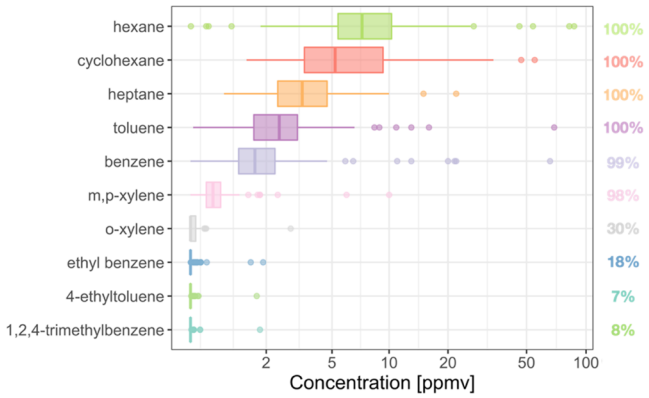


Figure 2. Summary of the top 10 non-methane volatile organic compounds (NMVOCs) detected in residential NG. Percent of values above the level of detection are listed on the right side of the figure. Note: the *x*-axis is a "pseudo-log" scale, which smoothly transitions to a linear scale around zero.

Median concentrations of BTEX were all within 0.1 ppmv of the means in the following regions: Greater Los Angeles, Bakersfield, Fresno, and Sacramento. This suggests regionally normal distributions of BTEX, further supported by a Kolmogorov−Smirnov test for benzene in these regions ($p > 0.05$; Figure 3 and Table S1). Notably, samples taken in the North San Fernando and Santa Clarita Valleys were not normally distributed and contained several very high benzene concentrations, including the maximum benzene concentration observed in the study—66 ppmv—which is approximately 66 times greater than the highest benzene level recorded in end-use NG in Massachusetts (max: 1.0 ppmv, mean: 0.17 ppmv).[24]

Benzene and BTEX by Gas Company. We compared the chemical composition of NG across all three major gas utility territories in California: PG&E, SoCalGas, and SDG&E (Figures 1, 3, S2, and S3 and Table S3). Total BTEX content differed significantly by gas company (Kruskal Wallace: $x^2 = 25.5$, $p < 0.0001$, $n = 3$), driven in part by the significantly lower BTEX observed in the SDG&E territory compared to PG&E ($p < 0.0001$) and SoCalGas ($p < 0.0001$) territories (Wilcoxon Rank Sum Test). Samples taken in SoCalGas service areas showed the highest variability and the highest overall concentrations—over 22% of samples in SoCalGas were above the maximum benzene concentrations of 3.8 ppmv from PG&E and 3.5 ppmv from SDG&E. Differences in benzene and BTEX concentrations between SoCalGas and SDG&E were particularly unexpected, given that SDG&E is a wholesale customer of SoCalGas and purchases NG directly from SoCalGas for distribution.[54,55] Further, both utilities reported sourcing NG from similar distribution pipeline systems and underground NG storage facilities (Note S2 and Figure S4), with the majority of gas originating from either the El Paso Pipelines sourced from basins in the Southwestern United States (i.e., the San Juan, Permian, or Anadarko Basins) or the Kern River Pipeline that sources NG from the Rocky Mountain Basin.[56] A fraction of gas comes from "California Sources," but we did not find publicly available data that specifies the exact source within the state (Figure S4). While BTEX concentrations did not significantly differ between SoCalGas and PG&E ($p = 0.12$), the vast majority of NG distributed by PG&E comes from Gas Transmission Northwestern pipeline, which sources NG from Western Canada,[56] indicating that while NG distributed by PG&E and SoCalGas is likely sourced from different basins, their C6+ content (NMVOCs with six or more carbons) was not significantly different.

Regional Benzene and BTEX Concentrations. Regional differences in BTEX concentrations (Kruskal Wallace: $x^2 = 46.7$, $p < 0.0001$, $n = 7$) were more pronounced than our observed differences by gas company. We observed exceptionally high NMVOCs collected in the North San Fernando and Santa Clarita Valleys (near the Aliso Canyon underground NG storage facility) and therefore analyzed these samples independently from the remainder of the samples in the Greater Los Angeles region. Notably, the 14 samples collected in this region—including 8 unique sites and 5 repeated samples in different months—exhibited some of the highest benzene and BTEX content observed throughout the entire study, with an overall mean benzene of 12 ppmv and a mean BTEX of 25 ppmv. We observed the maximum benzene concentration in the study (66 ppmv) in this region. The remaining samples collected in the Greater Los Angeles region had the most variability of the remaining regions, with a BTEX 95% CI of 7.5−24 ppmv, compared to the next highest BTEX 95% CI of 4.7−5.4 ppmv for the San Francisco Bay Area. Sacramento and Fresno had mean BTEX concentrations of 5.7 and 4.7 ppmv, respectively (Table S3). San Diego had relatively low BTEX concentrations compared to the other regions. Both Fresno and Bakersfield are located in the Central Valley but are serviced by different gas companies (PG&E and SoCalGas, respectively). This could partially account for their significantly lower BTEX content; however, these two areas represented the lowest sample counts with 12 and nine, respectively, and therefore required additional sampling to verify observed differences.

Overall, NMVOC and HAP concentrations reported in this study agree with the limited available data on NMVOC and HAP content observed in the midstream NG sector, including NG flowing through transmission pipelines, suggesting that the NG undergoes little additional processing.[25] For example, a study of HAPs present in transmission NG throughout the United States noted a mean benzene value of 5.9 ppmv (σ: ±6.0 ppmv), suggesting the composition of NG in transmission pipelines may not differ substantially from that of distribution gas at the point of end use.[25,57] Additionally, three preexisting NG composition samples associated with the Aliso Canyon underground NG storage facility (near Porter Ranch, CA) reported mol % benzene ranging from 0.0003−0.001 (3−10 ppmv),[58] which falls within the 25th−75th quartiles observed in samples in the North San Fernando and Santa Clarita Valleys from this study (1.8−20 ppmv).

While determining potential explanatory factors of trace gas variability was beyond the scope of this study, trace gas variability observed indicates that California's NG supply chain is complex and likely reflects the multiple hydrocarbon sources both from in-state production and imports in addition to the variability of the efficacy of NG processing systems that support end-use consumption. This is further evidenced by the five samples in the North San Fernando/Santa Clarita Valleys, where we sampled during different seasons. On average, the

https://doi.org/10.1021/acs.est.2c02581
Environ. Sci. Technol. 2022, 56, 15828−15838

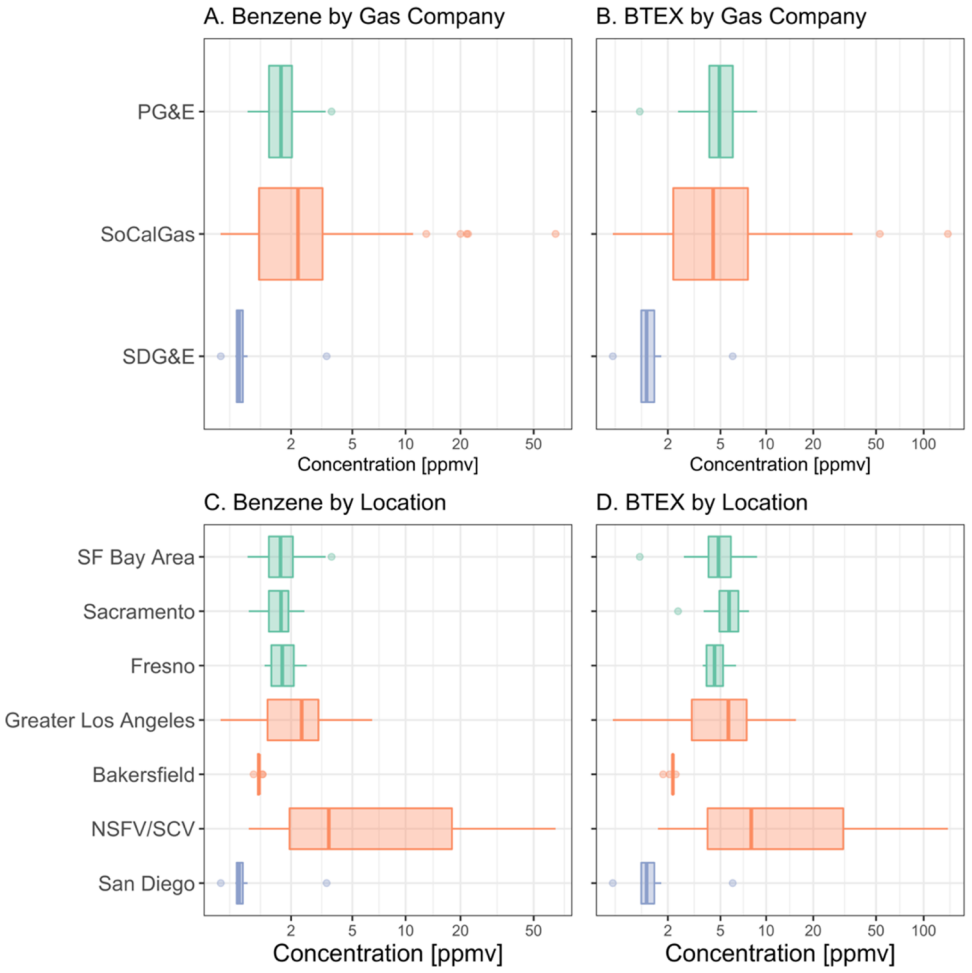


Figure 3. Concentrations of benzene and BTEX (sum of benzene, ethylbenzene, toluene, *m*,*p*-xylene, *o*-xylene) detected in residential NG by gas company and by location. Note that samples by location (C and D) are color-coded based on their respective gas company (A and B). Some samples collected in the North San Fernando and Santa Clarita Valleys (NSFV/SCV) had exceptionally high concentrations of BTEX in the gas; therefore, we analyzed these samples as a separate region from the remainder of the samples taken in the Greater Los Angeles region. No samples from the NSFV/SCV were included in the Greater Los Angeles region. Note: *x*-axis are a "pseudo-log" scale, which smoothly transitions to a linear scale around zero.

concentration of benzene in samples taken in the fall was 56% lower than samples taken in the winter/spring at the same location. Michanowicz also observed increased NMVOC concentrations during the winter heating season, suggesting that seasonal variability may be predictable and caused by source switching and/or influences from ambient temperature.[24]

Regional and Statewide Benzene and BTEX Emission Estimates. Assuming that the NMVOC content in unburned NG observed in our study is representative of California's transmission and distribution segments of the supply chain, we estimated annual statewide benzene and BTEX emissions using NG methane emission estimates of indoor appliances by Fischer et al.[26] and Lebel et al.[28] Our study suggests that there is an estimated 310 (95% CI: 140−980) kg benzene yr^{-1} emitted statewide from whole-house residential leaks, with an estimated 63 (95% CI: 32−150) kg benzene yr^{-1} emitted from gas stove steady-state-off leakage alone. For comparison, the 2017 NEI estimates benzene emissions from residential NG combustion (not unburned leakage) in California to be 381 kg benzene yr^{-1}.[59] Adding together benzene from combustion and our estimate of benzene from unburned leaks, our results imply that the California residential sector emits an estimated 690 kg benzene yr^{-1} from NG combustion and leakage. This suggests that the current emissions inventory underestimates benzene emissions for the residential sector by approximately 44%. This suggests that our estimate increases the existing inventory for benzene emissions from the residential sector by approximately 81%.

Using transmission and distribution methane estimates from Jeong et al.,[41] the results of our study indicate that an estimated 4,200 (95% CI: 1,800−9,700) kg benzene yr^{-1} is emitted from leaks in NG distribution and transmission systems across California (Table 1).[41] For context, this is a similar amount of benzene as emitted by 58,800 cars annually.[59,60] It is also equal to nearly half of the estimated benzene emissions (~47%) from all on-road diesel light-duty vehicles in California and would rank 24th out of 49 categories of California's benzene emissions tracked by the U.S. National Emissions Inventory (NEI).[59]

Given that statewide variability in BTEX concentrations is largely driven by the Greater Los Angeles Area, we also estimated regional benzene and BTEX emissions using regional NG emission estimates by Jeong et al.,[42] He et al.,[43] and SoCalGas[44] where available (Table 1). We estimated that 8.8 (95% CI: 4.5−19) kg benzene yr^{-1} is emitted from

https://doi.org/10.1021/acs.est.2c02581
Environ. Sci. Technol. 2022, 56, 15828−15838

Table 1. California Statewide and Regional Benzene and BTEX Emission Estimates Based on NG Methane Emissions from Previous Studies[a,b,c]

study	region	sector	methane emissions (Gg/yr) (95% CI)	implied benzene emissions (kg/yr) (95% CI)	implied BTEX emissions (kg/yr) (95% CI)
Jeong et al.[55]	Statewide	NG transmission and distribution emissions	317 (179−455)	4,200 (1,800−9,700)	11,000 (5,200−26,000), 500 (5,200−25,900)
Fischer et al.[25]	Statewide	residential appliances steady-state operation/combustion	13.3 (6.6−37.1)	170 (67−790)	480 (192−2,100)
	Statewide	residential whole-house quiescent leaks (i.e., pilot lights, pipes, etc.)	23.4 (13.7−45.6)	310 (140−980)	850 (400−2,600)
	Statewide	residential whole-house total emissions	35.7 (21.7−64.0)	470 (220−1,400)	1,300 (630−3,600)
Lebel et al.[27]	Statewide	residential NG stove steady-state-off leaks	4.8 (3.1−7.0)[d]	63 (32−150)	170 (90−400)
Jeong et al.[56]	San Francisco Bay Area	whole-region emissions (primarily attributable to distribution)	(23−38)[e]	(180−370)	(590−1,200)
He et al. 2019[57]	Greater Los Angeles	whole-region emissions based on residential and commercial NG consumption	72.8 (67.6−78.0)[f]	850 (590−1,100)	2,200 (1,600−3,000)
SoCalGas[58]	North San Fernando/ Santa Clarita Valleys	utility distribution emissions	0.15[g]	8.8 (4.5−19)	20 (9.5−50)

[a]BTEX emissions are calculated by summing the emissions of the individual compounds—benzene, toluene, ethylbenzene, and all xylenes. [b]95% confidence intervals are calculated by scaling the upper and lower values of the methane estimate from the reported study and the confidence interval of the concentration of the compound from this study. [c]Regional estimates were calculated using regional concentrations of compounds from this study. [d]Based on mean steady-state-off stove emissions of 57.9 (36.3−84; 95% CI) mg CH_4 hr^{-1}. [e]Based on the assumption that NG distribution emissions are proportional to consumption. Represents a leakage rate of 0.3−0.5%. [f]Based on residential and commercial NG consumption in the South Coast Air Basin in 2020 and a leakage rate of 1.4 ± 0.1%. [g]Distribution pipeline emissions in 2020 for zip codes within an ∼5 mile radius of Porter Ranch.

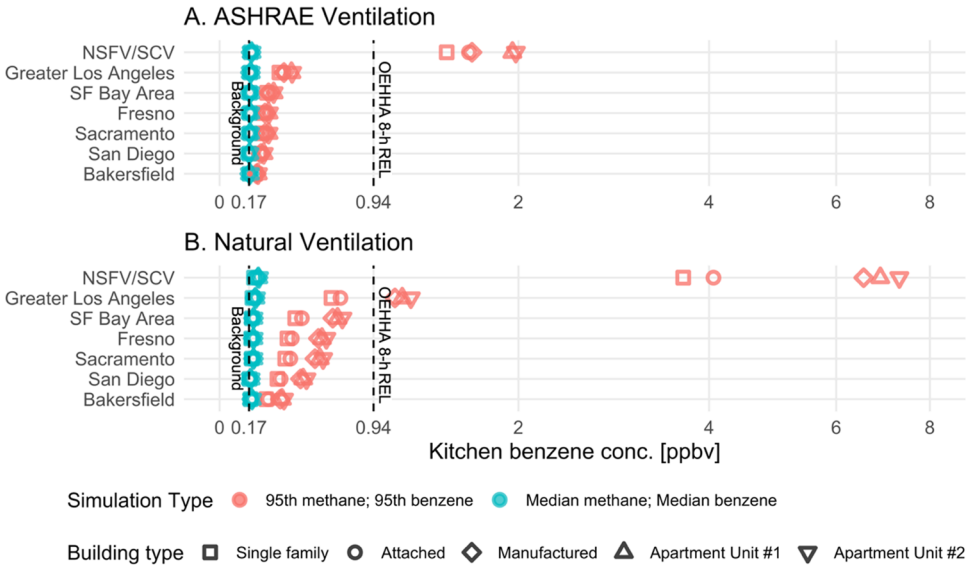


Figure 4. Kitchen indoor air benzene concentration estimates from steady-state-off CONTAM model simulations. Results are for two scenarios of ventilation: ASHRAE standard mechanical ventilation of 0.35 air changes per hour (ACH) and natural ventilation (0.05 to 0.11 ACH). The vertical dashed line shows the" California Environmental Protection Agency Office of Environmental Health Hazard Assessment (OEHHA) 8-h and chronic Reference Exposure Level (REL) for benzene (3 $\mu g\ m^{-3}$; 0.94 ppb). Median and 95th percentile methane emissions from gas stoves while off (24 and 282 mg hr^{-1}, respectively) are taken from Lebel et al.[28] Median and 95th percentile benzene concentrations are taken from this study (Figure 3C,D and Table S3). Building types are shown by various shapes and also have different-sized kitchens ranging from 29−33 m^3. Apartments 1 and 2 are the same except that Apartment 1 is on the ground floor and Apartment 2 is on the upper floor. Note: Greater Los Angeles does not include samples from the North San Fernando/Santa Clarita Valleys (NSFV/SCV).

SoCalGas distribution pipelines in the North San Fernando and Santa Clarita Valleys alone, where BTEX concentrations were the highest observed (Table S4). These estimates assume that the composition of NG for all sources is relatively stable and do not account for NMVOCs that may be produced or destroyed during the combustion process.[61−63]

While previous studies have estimated HAP emissions from NG production and development and their implications on air quality,[6,64−67] to our knowledge, HAP emissions from leaking downstream infrastructure and NG appliances are rarely measured and are not included in current inventories. Our regional estimates of benzene emissions in the San Francisco Bay Area (Table 1) are similar to what Michanowicz et al.[24] estimated in the Greater Boston Area (120−356 kg benzene yr^{-1}) but are 3−5 times higher in the Greater Los Angeles region.[24] Moreover, these inferred NMVOC emissions have likely been misclassified or unaccounted for as sources of

https://doi.org/10.1021/acs.est.2c02581
Environ. Sci. Technol. 2022, 56, 15828−15838

secondary formation of ozone and $PM_{2.5}$, particularly in nonattainment areas.

Modeled Benzene Kitchen Concentrations. Most model simulations—including all median value simulations—did not result in ambient benzene concentrations attributable to emissions of NG from gas stoves that are off above the California Environmental Protection Agency Office of Environmental Health Hazard Assessment (OEHHA) 8-h REL of 0.94 ppbv (also equal to the chronic REL). However, we found that certain parameter combinations have the potential to lead to exceedances of the OEHHA benzene REL from gas stoves while off (Figure 4).[37] Modeled indoor ambient benzene concentrations ranged from 38−126% of the 8-h REL in the Greater Los Angeles region and reached over 700% the REL in the North San Fernando/Santa Clarita Valleys. Notably, all 95th percentile model runs for the NSFV/SCV region resulted in REL exceedances irrespective of ventilation or building type or other indoor sources of benzene. These exceedances produce indoor concentrations of benzene similar to concentrations to environmental tobacco smoke (i.e., secondhand smoke).[68,69]

Based on model results, an elevated leakage rate of benzene and a low ventilation rate are both requisite for indoor concentrations to exceed the OEHHA 8-h REL for benzene. Using multiple linear regression, we found that the benzene emission rate (benzene concentration times methane flow rate) was the most important factor for determining the kitchen benzene concentration ($p < 0.0001$), followed by the air changes per hour ($p < 0.0001$)—a measure of the air tightness of the space. Interestingly, building size had an insignificant effect on benzene concentration ($p = 0.19$), possibly due to a small number of modeled home sizes. Modeled air changes per hour (ACH) for the natural ventilation simulations ranged from 0.11 for the single-family home to 0.05 for apartments, which has been verified empirically elsewhere (See the Supporting Information for more details).[70]

There are several reasons our estimates of indoor concentrations of benzene could underestimate true concentrations indoors. First, our calculations were exclusively focused on the contribution of benzene from NG leakage from gas stoves while off and do not include leakage from gas stoves during combustion or incomplete combustion,[62] other gas appliances (including but not limited to quiescent leakage from other appliances, see Table 1), gas pipes in residence, or other non-NG benzene sources. Second, we included a background ambient benzene concentration of 0.174 ppb for all model runs—the average outdoor ambient concentration in California in 2019.[71] This background benzene concentration may be a conservative measure particularly for some outdoor regions within our study area, and indoor VOCs are generally higher than outdoors to begin with. For example, the South Coast Air Quality Management District (SCAQMD) notes a background outdoor benzene concentration of 0.25 ppb.[72] We designed our modeling to identify a possible hazard (gas stoves while off only) and recognize that our parameters could lead to underestimating indoor benzene concentrations.

Future Research Directions. Our sampling effort was designed to capture geographic variability rather than temporal variability. While several of our samples were collected in different seasons, our study did not systematically investigate seasonal variability and its associated impacts (e.g., NG demand, withdrawal from underground NG storage facilities, etc.) on concentrations of NMVOCs in distribution gas. Future work could investigate temporal trends in California in greater depth, as temporal variability was evident in Boston.[24] Additionally, while we can infer the various high-volume transmission lines supplying NG regionally in California, we were unable to ultimately source NMVOC composition by upstream source or any potential modifying factors. The highly elevated NMVOC concentrations in distribution NG in the North San Fernando/Santa Clarita Valleys warrant further study to better understand factors contributing to these high concentrations.

Our CONTAM modeling only included emissions of unburned NG leaking from one source: stoves while off. Future work should consider air quality impacts and human health risks of exposure to multiple HAPs and other air pollutants (beyond benzene), emissions of unburned NG from appliances beyond stoves, and indoor emissions from burning NG to build a more complete picture of the potential indoor air quality and health impacts of household NG usage. Additionally, fugitive VOCs from downstream NG systems are likely an under-appreciated contributor to the secondary formation of $PM_{2.5}$ and ozone—contributing to outdoor air pollution—and should be studied further. As transportation and industry-related VOC emissions continue to decline in urban areas, NG-sourced VOCs may increase in their relative importance as $PM_{2.5}$ and ozone precursors.

Policy Implications. While there has been substantial research on the health cobenefits of greenhouse gas emission-reduction strategies in transportation, electricity generation, buildings, and industry that has helped inform policy, there is limited research on the health benefits of policies that reduce NG emissions and coemitted air pollutants. This is particularly true in urban areas with many emission sources and in indoor environments where people may be much more frequently exposed to NG-related emissions at higher concentrations. Thus, our study adds to the body of literature that suggests that downstream NG emissions are an unaccounted-for source of both indoor and outdoor emissions of NMVOCs and HAPs. California is already starting to take action to reduce urban pollution from NG: in September 2022, California air regulators approved a commitment to phasing out the sale of NG-fired furnaces and water heaters by 2030. The state has also moved to phase out subsidies for connecting new homes to the NG system, and is considering ending rebates for gas appliances to incentivize the transition to electric alternatives.[73,74]

In August 2021, the California Energy Commission adopted its 2022 Building Energy Efficiency Standards, which, in part, included differentiated demand-controlled ventilation requirements for electric stoves vs NG-fired stoves.[75] While this could lead to important improvements in indoor air quality related to combustion-related pollutants (e.g., NO_x, $PM_{2.5}$, CO_2), this ventilation requirement likely does not impact indoor air quality associated with fugitive, steady-state-off cooking appliance NG emissions—the source-exposure pathway characterized and modeled in this study.

While operating a continuous HVAC system would address nontransitory exposures, it is both cost- and energy-intensive, and retrofitting existing built environments like apartment complexes may not be feasible. Moreover, failing to address indoor pollution sources (e.g., cooking) has been shown to lead to unaccounted-for indoor air quality disbenefits following energy efficiency weatherization such as building sealing.[76] Our

https://doi.org/10.1021/acs.est.2c02581
Environ. Sci. Technol. 2022, 56, 15828−15838

study suggests that weatherization and building efficiency measures should be coupled with residential and other building electrification measures to support multiple health, air quality, and climate cobenefits by mitigating HAPs and methane emissions while appliances are both off and on.[77−81] In California, the California Public Utilities Commission (CPUC) pilot programs, TECH and BUILD, aim to reduce the use of NG in the home and provide incentives for all-electric new housing—with the latter program focusing on low-income households.[82,83] Given the potential air quality impacts and associated health risks from fugitive NG emissions alone—particularly those that may occur indoors—these electrification programs likely entail additional and unaccounted-for health cobenefits and associated long-term cost savings.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.2c02581.

Details on sample analysis, locations, and pollutant data; details on CONTAM modeling, and summary of public information on California NG sources and storage facilities (PDF)

■ AUTHOR INFORMATION

Corresponding Author

Eric D. Lebel − *PSE Healthy Energy, Oakland, California 94612, United States*; Email: elebel@psehealthyenergy.org

Authors

Drew R. Michanowicz − *PSE Healthy Energy, Oakland, California 94612, United States; Harvard T.H. Chan School of Public Health, C-CHANGE, Boston, Massachusetts 02215, United States;* orcid.org/0000-0002-2538-4819

Kelsey R. Bilsback − *PSE Healthy Energy, Oakland, California 94612, United States;* orcid.org/0000-0002-5996-1522

Lee Ann L. Hill − *PSE Healthy Energy, Oakland, California 94612, United States;* orcid.org/0000-0001-9919-9416

Jackson S. W. Goldman − *PSE Healthy Energy, Oakland, California 94612, United States;* orcid.org/0000-0002-9239-2982

Jeremy K. Domen − *PSE Healthy Energy, Oakland, California 94612, United States*

Jessie M. Jaeger − *PSE Healthy Energy, Oakland, California 94612, United States*

Angélica Ruiz − *PSE Healthy Energy, Oakland, California 94612, United States*

Seth B. C. Shonkoff − *PSE Healthy Energy, Oakland, California 94612, United States; Department of Environmental Health Sciences, School of Public Health, University of California, Berkeley, California 94720, United States; Energy Technologies Area, Lawrence Berkeley National Lab, Berkeley, California 94720, United States;* orcid.org/0000-0002-2696-0259

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.est.2c02581

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

The authors sincerely thank all of the study participants who volunteered their kitchen stoves for our study, especially in the midst of the COVID-19 pandemic. The authors especially thank all of the various organizations in California who shared this study with their members. The authors thank Molly Fairchild for sharing starting materials and strategies at the beginning of the project. We thank the Energy Foundation for providing funding for this study.

■ REFERENCES

(1) US EIAResidential Energy Consumption Survey (RECS): 2020 RECS Survey Data. https://www.eia.gov/consumption/residential/data/2020/#fueluses. (accessed July 06, 2022).

(2) US EIANatural Gas Explained: Use of Natural Gas. https://www.eia.gov/energyexplained/natural-gas/use-of-natural-gas.php. (accessed June 14, 2022).

(3) US EIANatural Gas Consumption by End Use. https://www.eia.gov/dnav/ng/ng_cons_sum_dcu_nus_a.htm. (accessed December 22, 2021).

(4) DiGiulio, D. C.; Jackson, R. B. Impact to Underground Sources of Drinking Water and Domestic Wells from Production Well Stimulation and Completion Practices in the Pavillion, Wyoming, Field. *Environ. Sci. Technol.* **2016**, *50*, 4524−4536.

(5) Faramawy, S.; Zaki, T.; Sakr, A.-E. Natural Gas Origin, Composition, and Processing: A Review. *J. Nat. Gas Sci. Eng.* **2016**, *34*, 34−54.

(6) Marrero, J. E.; Townsend-Small, A.; Lyon, D. R.; Tsai, T. R.; Meinardi, S.; Blake, D. R. Estimating Emissions of Toxic Hydrocarbons from Natural Gas Production Sites in the Barnett Shale Region of Northern Texas. *Environ. Sci. Technol.* **2016**, *50*, 10756−10764.

(7) Pétron, G.; Karion, A.; Sweeney, C.; Miller, B. R.; Montzka, S. A.; Frost, G. J.; Trainer, M.; Tans, P.; Andrews, A.; Kofler, J.; et al. A New Look at Methane and Nonmethane Hydrocarbon Emissions from Oil and Natural Gas Operations in the Colorado Denver-Julesburg Basin. *J. Geophys. Res.: Atmos.* **2014**, *119*, 6836−6852.

(8) US EPA. *Locating and Estimating Air Emissions from Sources of Benzene*; EPA-454/R-98-011; U.S. Environmental Protection Agency, 1998; p 361.

(9) Warneke, C.; Geiger, F.; Edwards, P. M.; Dube, W.; Pétron, G.; Kofler, J.; Zahn, A.; Brown, S. S.; Graus, M.; Gilman, J. B.; et al. Volatile Organic Compound Emissions from the Oil and Natural Gas Industry in the Uintah Basin, Utah: Oil and Gas Well Pad Emissions Compared to Ambient Air Composition. *Atmos. Chem. Phys.* **2014**, *14*, 10977−10988.

(10) Liss, W. E.; Thrasher, W. H.; Steinmetz, G. F.; Chowdiah, P.; Attari, A.*Variability of Natural Gas Composition in Select Major Metropolitan Areas of the United States*, Final Report, August 1990−February 1992, 1992.

(11) European Commission. Commission Regulation (EU) 2015/1494 of 4 September 2015 Amending Annex XVII to Regulation (EC) No 1907/2006 of the European Parliament and of the Council Concerning the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH) as Regards Benzene. *Off. J. Eur. Union* **2015**, *58*, No. 2.

(12) *Policy Statement on Provisions Governing Natural Gas Quality and Interchangeability in Interstate Natural Gas Pipeline Company Tariffs*, Docket No. PL04-3-000; Federal Energy Regulatory Commission, 2006.

(13) *FERC Gas Tariff - Third Revised Volume No. 1*; Kern River Gas Transmission Company, 2016.

(14) PG&EGas Rule No. 21 - Transportation of Gas2020.

(15) SoCalGasRule No. 30 - Transportation of Customer-Owned Gas,2011.

(16) Texas Eastern Transmission, LP. FERC Gas Tariff - Eighth Revised Volume No. 1 of Texas Eastern Transmission, LP2017.

(17) *Chemical Agents and Related Occupations*, IARC Monographs on the Evaluation of 532 Carcinogenic Risk to Humans; International Agency for Research on Cancer, 2012.
(18) WHO. *Air Quality Guidelines for Europe, Second Edition*; World Health Organization, 2000.
(19) USEPA. *Technical Overview of Volatile Organic Compounds*.
(20) Jimenez, J. L.; Canagaratna, M. R.; Donahue, N. M.; Prevot, A. S. H.; Zhang, Q.; Kroll, J. H.; DeCarlo, P. F.; Allan, J. D.; Coe, H.; Ng, N. L.; Aiken, A. C.; Docherty, K. S.; Ulbrich, I. M.; Grieshop, A. P.; Robinson, A. L.; Duplissy, J.; Smith, J. D.; Wilson, K. R.; Lanz, V. A.; Hueglin, C.; Sun, Y. L.; Tian, J.; Laaksonen, A.; Raatikainen, T.; Rautiainen, J.; Vaattovaara, P.; Ehn, M.; Kulmala, M.; Tomlinson, J. M.; Collins, D. R.; Cubison, M. J.; E; Dunlea, J.; Huffman, J. A.; Onasch, T. B.; Alfarra, M. R.; Williams, P. I.; Bower, K.; Kondo, Y.; Schneider, J.; Drewnick, F.; Borrmann, S.; Weimer, S.; Demerjian, K.; Salcedo, D.; Cottrell, L.; Griffin, R.; Takami, A.; Miyoshi, T.; Hatakeyama, S.; Shimono, A.; Sun, J. Y.; Zhang, Y. M.; Dzepina, K.; Kimmel, J. R.; Sueper, D.; Jayne, J. T.; Herndon, S. C.; Trimborn, A. M.; Williams, L. R.; Wood, E. C.; Middlebrook, A. M.; Kolb, C. E.; Baltensperger, U.; Worsnop, D. R. Evolution of Organic Aerosols in the Atmosphere. *Science* **2009**, *326*, 1525−1529.
(21) McDonald, B. C.; de Gouw, J. A.; Gilman, J. B.; Jathar, S. H.; Akherati, A.; Cappa, C. D.; Jimenez, J. L.; Lee-Taylor, J.; Hayes, P. L.; McKeen, S. A.; Cui, Y. Y.; Kim, S.-W.; Gentner, D. R.; Isaacman-VanWertz, G.; Goldstein, A. H.; Harley, R. A.; Frost, G. J.; Roberts, J. M.; Ryerson, T. B.; Trainer, M. Volatile Chemical Products Emerging as Largest Petrochemical Source of Urban Organic Emissions. *Science* **2018**, *359*, 760−764.
(22) USEPA. Benzene.
(23) Burger, J. L.; Lovestead, T. M.; Bruno, T. J. Composition of the C6+ Fraction of Natural Gas by Multiple Porous Layer Open Tubular Capillaries Maintained at Low Temperatures. *Energy Fuels* **2016**, *30*, 2119−2126.
(24) Michanowicz, D. R.; Dayalu, A.; Nordgaard, C. L.; Buonocore, J. J.; Fairchild, M. W.; Ackley, R.; Schiff, J. E.; Liu, A.; Phillips, N. G.; Schulman, A.; Magavi, Z.; Spengler, J. D. Home Is Where the Pipeline Ends: Characterization of Volatile Organic Compounds Present in Natural Gas at the Point of the Residential End User. *Environ. Sci. Technol.* **2022**, *56*, 10258−10268.
(25) Nordgaard, C. L.; Jaeger, J. M.; Goldman, J.; Shonkoff, S. B. C.; Michanowicz, D. R. Contamination of Transmission Pipeline Natural Gas by Hazardous Air Pollutants: A Systematic Analysis of Industry Disclosures and Data. *Environ. Res. Lett.* **2022**, *17*, 104032.
(26) Fischer, M. L.; Chan, W. R.; Delp, W.; Jeong, S.; Rapp, V.; Zhu, Z. An Estimate of Natural Gas Methane Emissions from California Homes. *Environ. Sci. Technol.* **2018**, *52*, 10205−10213.
(27) Lebel, E. D.; Lu, H. S.; Speizer, S. A.; Finnegan, C. J.; Jackson, R. B. Quantifying Methane Emissions from Natural Gas Water Heaters. *Environ. Sci. Technol.* **2020**, *54*, 5737−5745.
(28) Lebel, E. D.; Finnegan, C. J.; Ouyang, Z.; Jackson, R. B. Methane and NO $_x$ Emissions from Natural Gas Stoves, Cooktops, and Ovens in Residential Homes. *Environ. Sci. Technol.* **2022**, *56*, 2529−2539.
(29) Crippen, K.; Hill, A.; Baffoe, J.; Glanville, P.*Odorant Effectiveness*; DOT Prj# 363, Contract Number: DTPH56-10-T-000018; Gas Technology Institute, 2013.
(30) Dalton, P. Psychophysical and Behavioral Characteristics of Olfactory Adaptation. *Chem. Senses* **2000**, *25*, 487−492.
(31) Larsson, M.; Finkel, D.; Pedersen, N. L. Odor Identification: Influences of Age, Gender, Cognition, and Personality. *J. Gerontol., Ser. A* **2000**, *55*, P304−P310.
(32) Matsubasa, T.; Gotow, N.; Gomi, Y.; Kobayakawa, T. A Method for Psychophysical Screening of Odorants for Use in City Gas Based on Olfactory Adaptation Tolerance. *Chemosens. Percept.* **2016**, *9*, 120−130.
(33) Moschandreas, D.; Jones, D.; Martinus, J.; Ragus, E.*Odor Recognition and Fatigue*; IIT Research Inst.: Chicago, IL (USA), 1982.
(34) Wise, P.; Rowe, S.; Dalton, P.*Odorization of Natural Gas: What Are the Challenges?*, 2021, DOI: 10.1101/2021.07.10.450231.
(35) Dols, W. S.; Polidoro, B. J.*CONTAM User Guide and Program Documentation Version 3.4*, NIST TN 1887 Rev. 1; National Institute of Standards and Technology, 2020.
(36) Dols, W. S.; Emmerich, S. J.; Polidoro, B. J. Using Coupled Energy, Airflow and Indoor Air Quality Software (TRNSYS/CONTAM) to Evaluate Building Ventilation Strategies. *Build. Serv. Eng. Res. Technol.* **2016**, *37*, 163−175.
(37) OEHHA. OEHHA Acute, 8-h and Chronic Reference Exposure Level (REL) Summary. https://oehha.ca.gov/air/general-info/oehha-acute-8-hour-and-chronic-reference-exposure-level-rel-summary. (accessed Jull 19, 2022).
(38) *Compendium of Methods for the Determination of Toxic Organic Compounds in Ambient Air*, 2nd ed.; U.S. Environmental Protection Agency: Cincinnati, OH, 1999.
(39) *Standard Practice for Analysis of Reformed Gas by Gas Chromatography*, D1946-90R19; ASTM, 2019.
(40) US EPA. Regional Guidance on Handling Chemical Concentration Data Near the Detection Limit in Risk Assessments: Regional Technical Guidance Manual, Risk Assessment. https://www.epa.gov/risk/regional-guidance-handling-chemical-concentration-data-near-detection-limit-risk-assessments. (accessed July 19, 2022).
(41) Jeong, S.; Millstein, D.; Fischer, M. L. Spatially Explicit Methane Emissions from Petroleum Production and the Natural Gas System in California. *Environ. Sci. Technol.* **2014**, *48*, 5982−5990.
(42) Jeong, S.; Cui, X.; Blake, D. R.; Miller, B.; Montzka, S. A.; Andrews, A.; Guha, A.; Martien, P.; Bambha, R. P.; LaFranchi, B.; et al. Estimating Methane Emissions from Biological and Fossil-Fuel Sources in the San Francisco Bay Area. *Geophys. Res. Lett.* **2017**, *44*, 486−495.
(43) He, L.; Zeng, Z.-C.; Pongetti, T. J.; Wong, C.; Liang, J.; Gurney, K. R.; Newman, S.; Yadav, V.; Verhulst, K.; Miller, C. E.; et al. Atmospheric Methane Emissions Correlate with Natural Gas Consumption from Residential and Commercial Sectors in Los Angeles. *Geophys. Res. Lett.* **2019**, *46*, 8563−8571.
(44) *SoCalGas. Distribution Pipeline Emissions Map*.
(45) Dols, W. S.; Polidoro, B. J.*CONTAM User Guide and Program Documentation Version 3.2*, NIST TN 1887; National Institute of Standards and Technology, 2015.
(46) Palmgren, C.; Ramirez, B.; Goldberg, M.; Williamson, C.*2019 California Residential Appliance Saturation Study (RASS)*, CEC-200-2021-005; California Energy Commission; DNV GL Energy Insights USA, Inc, 2021.
(47) *Residential Appliance Saturation Study (RASS)*; CEC-200-2021-005; Prepared for California Energy Commission; DNV GL Energy Insights USA, Inc, 2021.
(48) Persily, A.; Leber, D.*NIST Interagency/Internal Report (NISTIR) 7330*, A Suite Of Homes Representing The U.s. Housing Stock; National Institute of Standards and Technology: Gaithersburg, MD; 2008.
(49) Seinfeld, J. H.; Pandis, S. N.*Atmospheric Chemistry and Physics: From Air Pollution to Climate Change*, 2nd ed.; John Wiley & Sons, Inc.: Hoboken, NJ, 2006.
(50) Weschler, C. J.; Shields, H. C. Production of the Hydroxyl Radical in Indoor Air. *Environ. Sci. Technol.* **1996**, *30*, 3250−3258.
(51) Singer, B. C.; Revzan, K. L.; Hotchi, T.; Hodgson, A. T.; Brown, N. J. Sorption of Organic Gases in a Furnished Room. *Atmos. Environ.* **2004**, *38*, 2483−2494.
(52) Singer, B. C.; Hodgson, A. T.; Hotchi, T.; Ming, K. Y.; Sextro, R. G.; Wood, E. E.; Brown, N. J. Sorption of Organic Gases in Residential Rooms. *Atmos. Environ.* **2007**, *41*, 3251−3265.
(53) US EPA. Health Effects Notebook for Hazardous Air Pollutants.. https://www.epa.gov/haps/health-effects-notebook-hazardous-air-pollutants. (accessed November 05, 2021).
(54) Sempra Energy. *Innovation, Sustainability, and Leadership 2020 Annual Report; 2020 Form 10-K*, 2021.
(55) *Southwest Gas Corporation; Long Beach Energy Resources Department*, California Gas Report; D.95-01-0392020; Southern California Edison Company, 2020.

(56) *California Gas Report Supplement*, D.95-01-039; Long Beach Energy Resources Department, 2021.
(57) *Composition of Natural Gas for Use in the Oil and Natural Gas Sector Rulemaking*; USEPA Docket EPA-HQ-OAR-2021-0317-0014; EC/R Inc, 2011.
(58) Terry, D.*Natural Gas Analyses: Chain of Custody 84422*; Texas Oil Tech Laboratories, Inc., 2017.
(59) US EPA.. National Emissions Inventory (NEI) Data, 2017. https://www.epa.gov/air-emissions-inventories/2017-national-emissions-inventory-nei-data#datas. (accessed July 19, 2022).
(60) California Energy Commission. Light-Duty Vehicle Population. https://www.energy.ca.gov/data-reports/energy-almanac/zero-emission-vehicle-and-infrastructure-statistics/light-duty-vehicle. (accessed September 29, 2022).
(61) Jin, H.; Frassoldati, A.; Wang, Y.; Zhang, X.; Zeng, M.; Li, Y.; Qi, F.; Cuoci, A.; Faravelli, T. Kinetic Modeling Study of Benzene and PAH Formation in Laminar Methane Flames. *Combust. Flame* **2015**, *162*, 1692−1711.
(62) Senkan, S.; Castaldi, M. Formation of Polycyclic Aromatic Hydrocarbons (PAH) in Methane Combustion: Comparative New Results from Premixed Flames. *Combust. Flame* **1996**, *107*, 141−150.
(63) Slavinskaya, N. A.; Frank, P. A Modelling Study of Aromatic Soot Precursors Formation in Laminar Methane and Ethene Flames. *Combust. Flame* **2009**, *156*, 1705−1722.
(64) Fann, N.; Baker, K. R.; Chan, E. A.; Eyth, A.; Macpherson, A.; Miller, E.; Snyder, J. Assessing Human Health PM2. 5 and Ozone Impacts from US Oil and Natural Gas Sector Emissions in 2025. *Environ. Sci. Technol.* **2018**, *52*, 8095−8103.
(65) Francoeur, C. B.; McDonald, B. C.; Gilman, J. B.; Zarzana, K. J.; Dix, B.; Brown, S. S.; de Gouw, J. A.; Frost, G. J.; Li, M.; McKeen, S. A.; Peischl, J.; Pollack, I. B.; Ryerson, T. B.; Thompson, C.; Warneke, C.; Trainer, M. Quantifying Methane and Ozone Precursor Emissions from Oil and Gas Production Regions across the Contiguous US. *Environ. Sci. Technol.* **2021**, *55*, 9129−9139.
(66) Halliday, H. S.; Thompson, A. M.; Wisthaler, A.; Blake, D. R.; Hornbrook, R. S.; Mikoviny, T.; Müller, M.; Eichler, P.; Apel, E. C.; Hills, A. J. Atmospheric Benzene Observations from Oil and Gas Production in the Denver-Julesburg Basin in July and August 2014. *J. Geophys. Res.: Atmos.* **2016**, *121*, 11055−11074.
(67) Hendryx, M.; Luo, J. Natural Gas Pipeline Compressor Stations: VOC Emissions and Mortality Rates. *Extr. Ind. Soc.* **2020**, *7*, 864−869.
(68) Singer, B. C.; Hodgson, A. T.; Nazaroff, W. W. Gas-Phase Organics in Environmental Tobacco Smoke: 2. Exposure-Relevant Emission Factors and Indirect Exposures from Habitual Smoking. *Atmos. Environ.* **2003**, *37*, 5551−5561.
(69) Nazaroff, W. W.; Singer, B. C. Inhalation of Hazardous Air Pollutants from Environmental Tobacco Smoke in US Residences. *J. Exposure Sci. Environ. Epidemiol.* **2004**, *14*, S71−S77.
(70) Huangfu, Y.; Lima, N. M.; O'Keeffe, P. T.; Kirk, W. M.; Lamb, B. K.; Walden, V. P.; Jobson, B. T. Whole-House Emission Rates and Loss Coefficients of Formaldehyde and Other Volatile Organic Compounds as a Function of the Air Change Rate. *Environ. Sci. Technol.* **2020**, *54*, 2143−2151.
(71) California Air Resources Board. Annual Statewide Toxics Summary: Benzene. https://www.arb.ca.gov/adam/toxics/statepages/benzstate.html. (accessed July 19, 2022).
(72) South Coast Air Quality Management District. *MATES V Multiple Air Toxics Exposure Study in the South Coast AQMD*; 2021.
(73) California Air Resources Board*Proposed State SIP Strategy*, 2022; 181.
(74) Adegbesan, A.*California Moves to Ban Natural Gas Furnaces and Heaters by 2030. Los Angeles Times*.https://www.latimes.com/business/story/2022-09-23/california-moves-to-ban-natural-gas-furnaces-and-heaters-by-2030. (accessed September 30, 2022).
(75) California Energy Commission. 2022 Building Energy Efficiency Standards Summary, 2021.
(76) Underhill, L. J.; Dols, W. S.; Lee, S. K.; Fabian, M. P.; Levy, J. I. Quantifying the Impact of Housing Interventions on Indoor Air Quality and Energy Consumption Using Coupled Simulation Models. *J. Exposure Sci. Environ. Epidemiol.* **2020**, *30*, 436−447.
(77) Chan, W. R.; Kim, Y.-S.; Less, B. D.; Singer, B. C.; Walker, I. S.*Ventilation and Indoor Air Quality in New California Homes with Gas Appliances and Mechanical Ventilation*; Lawrence Berkeley National Lab: Berkeley, CA, 2019.
(78) Logue, J. M.; Klepeis, N. E.; Lobscheid, A. B.; Singer, B. C. Pollutant Exposures from Natural Gas Cooking Burners: A Simulation-Based Assessment for Southern California. *Environ. Health Perspect.* **2014**, *122*, 43−50.
(79) Singer, B. C.; Pass, R. Z.; Delp, W. W.; Lorenzetti, D. M.; Maddalena, R. L. Pollutant Concentrations and Emission Rates from Natural Gas Cooking Burners without and with Range Hood Exhaust in Nine California Homes. *Build. Environ.* **2017**, *122*, 215−229.
(80) Zhao, H.; Chan, W. R.; Cohn, S.; Delp, W. W.; Walker, I. S.; Singer, B. C. Indoor Air Quality in New and Renovated Low-Income Apartments with Mechanical Ventilation and Natural Gas Cooking in California. *Indoor Air* **2021**, *31*, 717−729.
(81) Zhu, Y.; Connolly, R.; Lin, Y.; Mathews, T.; Wang, Z.*Effects of Residential Gas Appliances on Indoor and Outdoor Air Quality and Public Health*; UCLA Fielding School of Public Health: California, 2020.
(82) California Energy Commission. Building Initiative for Low-Emissions Development Program. https://www.energy.ca.gov/programs-and-topics/programs/building-initiative-low-emissions-development-program. (accessed July 19, 2022).
(83) California Public Utilities Commission*Decision Establishing Building Decarbonization Pilot Programs; Decision 20-03-027, R.19-01-011*, **2020**.

https://doi.org/10.1021/acs.est.2c02581
Environ. Sci. Technol. 2022, 56, 15828−15838