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Article

Inequality in Hazardous Air Pollutant Emissions and Concentrations Measured Over Los Angeles

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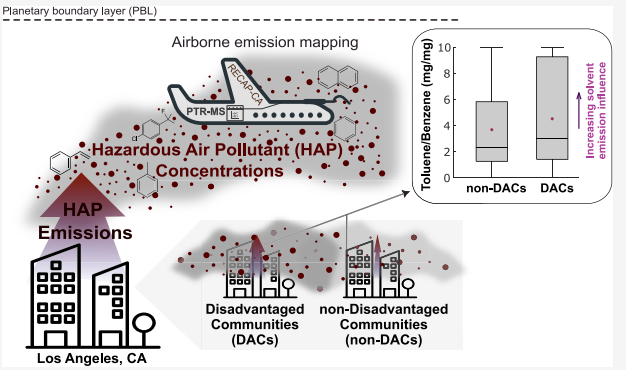
ABSTRACT: In Los Angeles, air pollution disproportionately impacts communities of color and low-income residents. Routine city-wide measurements of hazardous air pollutants (HAPs), of concern for health and contributing to urban air pollution, are notably lacking. In this study, we use the highest spatially resolved (∼2 km) measurements of emissions and concentrations ever reported of HAPs while covering a whole megacity and combine observations with US Census information. We observe higher concentrations and emissions of 17 measured HAPs, such as benzene, naphthalene, and *p*-chlorobenzotrifluoride (PCBTF), in California-designated Disadvantaged Communities (DACs) and census tracts with low-income Hispanics and Asians. These groups share an unequal burden from traffic-related emissions, with benzene, nitrogen oxides (NO_x), and carbon monoxide (CO) concentrations up to 60% higher. However, in DACs and census tracts with large Hispanic populations (>50%), we observe toluene-to-benzene emission ratios above 3, pointing to inequalities in other HAPs primarily caused by non-traffic emission sources such as industry and solvents. In these communities, regulatory inventories also significantly underestimate emissions. We find that efforts to address HAP inequalities and environmental justice concerns in Los Angeles will need to consider contributions from volatile chemical products, which represent a growing source of emissions driving inequalities in impacted communities.



KEYWORDS: *hazardous air pollutants (HAPs), environmental inequality, spatial distribution, HAP emission sources, Los Angeles*

1. INTRODUCTION

California's metropolitan Los Angeles (LA) is the second largest urban area in the United States, consistently ranking among the most polluted cities in the country.[1] The lasting effects of discriminatory practices such as redlining and racial segregation in LA[2] have contributed to the region's degraded air quality, disproportionately impacting low-income residents and communities of color.[3,4] These populations are also impacted by widespread emissions of hazardous air pollutants (HAPs), which have been linked to adverse health effects such as increased cancer risk, reproductive complications, developmental issues, and other serious health problems.[5−7]

Research has shown that HAP (e.g., benzene) emissions attributed mostly to transportation and small area sources in LA result in higher cancer risk for communities of color even after controlling for well-known explanatory factors of pollution such as income.[8] In addition, these communities tend to reside in closer proximity to industrial areas and hazardous sites in the region,[8,9] which contribute further to HAP emissions. City-wide measurements of HAP emissions and concentrations in LA are lacking compared to routinely investigated air pollutants such as fine particulate matter ($PM_{2.5}$) and nitrogen oxides (NO_x).[10−17] This lack of spatial information on HAP emissions arises from challenges associated with both direct measurements and air quality modeling.[18]

Unlike for $PM_{2.5}$ and NO_x, there are no low-cost sensors available for organic HAPs that would provide a high spatial resolution. Additionally, direct satellite observations of HAPs are currently not available, with the exception of formaldehyde. Mobile laboratories, while capable of high spatial resolution, would be strongly impacted by traffic emissions and are limited in their ability to directly measure emissions, as this requires sampling from a certain height above the atmospheric roughness layer. As a result, ongoing HAP monitoring efforts in LA focus on compliance of stationary industrial sources and sampling at sparse individual sites through regulatory ambient

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air quality monitoring stations or during field deployments of research studies.[19−22]

For instance, the South Coast Air Quality Management District (AQMD) conducts monitoring of HAPs across 10 sites in the LA region as part of the Multiple Air Toxics Exposure Study (MATES), with the most recent data spanning the 2018−2019 study period.[23] The US Environmental Protection Agency (EPA) also consolidates information on HAPs through different programs such as the Toxics Release Inventory, which serves as an input into the National Emissions Inventory (NEI). Through the NEI, the EPA models and quantifies HAP emissions, concentrations, and health risks through the Air Toxics Screening Assessment, with the most recent census tract-level data available from 2019.[24]

While regulatory efforts provide insight into the HAP landscape in LA, they incorporate indirect monitoring methods used to estimate the composition of HAP emissions, such as bottom-up emission inventories[25] or top-down inference of concentration measurements via chemical transport models.[26] These methods often rely on a range of assumptions, which can result in large uncertainties in HAP emissions.[18,27] Additionally, meteorology introduces further uncertainty by influencing the distribution of HAPs and the extent to which emissions translate into measurable concentrations, potentially impacting assessments of environmental inequalities.[28]

Spatially resolved atmospheric measurements of HAPs in LA are needed for assessing the local air quality impacts on the region's low-income residents and communities of color who are already overburdened.[29] Without direct measurements and an assessment of the influence of meteorology on the distribution of HAP emissions and concentrations in the region, efforts aimed at mitigating disparities and reducing overall health risks could be misallocated. To address this need, we employed state-of-the-art instrumentation[30] to perform in situ airborne eddy covariance measurements over the LA Basin, resulting in the first direct observations of spatially resolved emissions and concentrations of volatile organic compounds (VOCs) and other gaseous air pollutants in the region.

In this study, we focus on VOCs identified as HAPs by the EPA,[31] with direct measurements representing the highest spatial resolution (∼2 km) ever reported covering a megacity.[32] We combine a breadth of spatially resolved HAP observations with census tract-level information and quantify inequalities, specifically through race, ethnicity, and income. We discuss differences between population-weighted concentrations and emissions of HAPs, assess the spatial distribution of attributable emission sources, and compare direct observations to emission inventory estimates in relation to population groups. Overall, we highlight pertinent environmental justice concerns associated with the spatial distribution and inequality of HAPs and their implications in urban Los Angeles.

2. METHODS

2.1. Airborne Trace Gas Measurements and Climatology. Airborne measurements of VOCs, VOC fluxes, NO_x, and CO were conducted as part of the RECAP-CA (RE-evaluating chemistry of air pollutants in California) campaign. Nine out of 16 flights, all starting at the airport in Burbank, CA, were performed over the LA Basin and are the subject of this study (see flight tracks in Figure 1). The nine LA flights were conducted between 11:00 and 17:00 local time on different

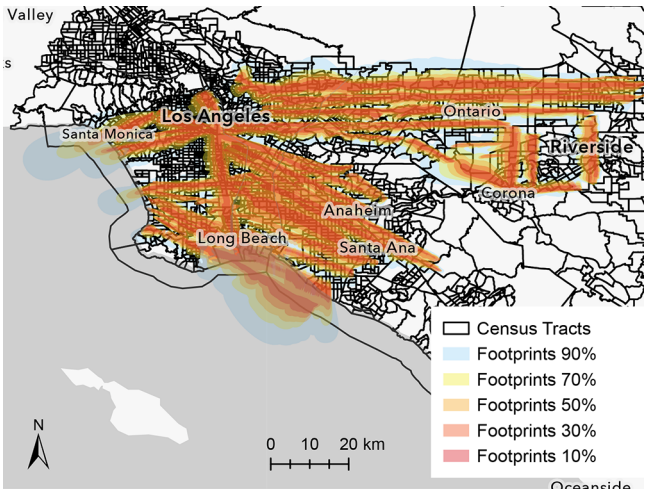

Figure 1. Map of the LA study area showing census tract contours, flight tracks, and flux footprints.

days between June 1 and 21, 2021, and details on flight meteorological conditions can be found in Table S1.

Detailed descriptions of the direct measurements and instrumentation, the aircraft, and wavelet transformation flux calculations can be found elsewhere for VOCs,[27] NO_x,[33] and CO.[34] Briefly, air was drawn into an isokinetic inlet above the nose of the campaign aircraft and subsampled in the aircraft cabin into each of the instruments for VOCs, NO_x, and CO. The original time resolutions of 0.1 s (VOCs), 0.2 s (NO_x), and 0.7 s (CO) were averaged to a spatial resolution of 200 m.

NO_x was measured by Laser-Induced Fluorescence[33] and CO by a cavity ring-down spectroscope (Picarro Inc.).[34] VOCs were measured by Vocus proton transfer reaction time-of-flight mass spectrometry (Vocus PTR-ToF-MS, Aerodyne Research Inc.).[30] PTR-ToF-MS allows for simultaneous observations of hundreds of VOCs at a high time resolution. Exact masses (m/z) derived from the observations can be attributed to chemical formulas that may represent one or more isomers of the same chemical composition. For this study, we focused on observed m/z corresponding to VOCs listed as HAPs by the EPA,[31] which are as follows: acetamide, acetophenone, acetaldehyde*, acetonitrile*, acrolein*, acrylonitrile*, benzene*, dichlorobenzene, dibenzofuran, nitrophenol, naphthalene, PCBTF, phenol, quinoline, styrene, toluene*, and xylene*. The compounds marked with an asterisk ("*") were calibrated with a gas standard and are therefore subject to low uncertainty (∼20% in concentrations, ∼75−86% in emission fluxes). For the remaining compounds, the sensitivities were derived from a commonly used theoretical calibration that accounts for transmission dependent on m/z.[35,36] The estimated uncertainty for these compounds is 54% in concentrations and 90−170% in emission fluxes. Known fragmentation and interferences in PTR-ToF-MS were accounted for. Acetaldehyde was corrected for the interference of ethanol fragmentation, benzene was calibrated on m/z = 78 to avoid the interference of benzaldehyde and ethylbenzene fragments,[37] and all identified fragments of PCBTF were added up to derive the PCBTF mixing ratios and fluxes.

2.2. Demographic Data and Disadvantaged Communities. The analyses in this study focus on race-ethnicity and income as these indicators are strong proxies for air pollution disparities,[38−40] with historical relevance and importance for the distribution of demographic groups in LA

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(see Text S1). Census tract-level demographic data were obtained from the US Census Bureau 2017−2021 American Community Survey (ACS).[41] Race-ethnicity demographics were defined with the following US Census codes: non-Hispanic white (JMJE003), Black/African American (JMJE004), and Asian (JMJE006), all excluding individuals identifying as Hispanic or Latino, and Hispanic/Latino (JMJE012), including all races reporting as Hispanic or Latino. Additionally, we performed isolation index calculations[42] to assess the spatial distribution of demographic groups in the LA study area. The isolation index measures the extent to which minority populations are exposed to only one another and allows us to observe trends in HAP observations for the studied groups.

Household income data were separated into low- and high-income census tracts. Low- and high-income census tracts are defined as census tracts below the 25th percentile and above the 75th percentile of median household incomes (MHI) in the LA Basin, respectively—i.e., <50,000 USD are low-income census tracts and >95,000 USD are high-income census tracts.

Disadvantaged Communities are defined from designated census tracts by CalEnviroScreen 4.0.[43] CalEnviroScreen 4.0 identifies communities in California disproportionately burdened by pollution with characteristic population vulnerabilities. This study utilizes CalEnviroScreen over alternative screening tools, such as the Climate and Economic Justice Screening Tool (CEJST), due to its curation and continuous development to provide the most accurate information on California's DACs. CalEnviroScreen 4.0 is also relevant for air pollution agencies in California that utilize the screening tool to adequately implement regulatory policies (e.g., Assembly Bill (AB) 617) that support DACs. In this study, only CalEnviroScreen 4.0 "Top 25%" and "High Pollution Burden Score, Low Population Count" census tracts were considered.

2.3. Population-Weighted Observations and Census Tract Inequalities. Population-weighted HAP mixing ratios and emissions (Data S1) were calculated using eq 1,[44] where c_j is the population-weighted concentration (or emission) of demographic group j, c_i is the concentration (or emission) in census tract i, $p_{i,j}$ is the population of demographic group j in census tract i, and n is the total number of sampled census tracts.

$$c_j = \frac{\sum_{i=1}^{n} c_i \times p_{i,j}}{\sum_{i=1}^{n} p_{i,j}} \quad (1)$$

Inequalities in mixing ratios (or emissions) are discussed in terms of relative (%) or absolute (ppb or mg m^{-2} h^{-1}) differences in population-weighted values between two demographic groups across sampled census tracts. Accordingly, discussed inequalities for Asians and Hispanics living in low-income census tracts are calculated in reference to non-Hispanic whites living in high-income census tracts. Inequalities for Disadvantaged Communities use community-averaged mixing ratios (or emissions) in reference to non-Disadvantaged Communities for the sampled area (Figure S1).

2.4. Matching Airborne Observations with Census Tracts and Inventory Grids. During all flights, the flight altitude was held constant at ∼350−400 ± 50 m. The flight times during daytime ensured a well-mixed, high boundary layer, which allowed the aircraft to fly inside the boundary layer. Boundary layer heights were estimated based on stark drops in water concentration, dewpoint temperature, and VOC concentrations during soundings performed at the beginning, during, and end of each flight.[27] Thus, we were able to identify data points measured outside the boundary layer and remove them from the analysis. The mixing ratios of relatively long-lived trace gases measured at 350−400 m inside the well-mixed boundary layer, such as the hazardous air pollutants discussed in this study, can be expected to correspond to the mixing ratios at the surface.[45] This is reflected in comparable HAP concentrations from prior ground-based studies and measurements in the LA region.[20−23] Table S2 shows ground-level HAP concentration data from MATES V,[23] which scales with our study observations (Data S1).

To estimate the atmospheric concentrations experienced by a demographic group or community in the respective census tracts, the mixing ratios were matched to the census tract below each point of observation. Census tracts with less than three observations, corresponding to 60 data points at the original VOC time resolution, were not considered. Different from concentrations, each flux data point measured at the flight altitude (in units of emission per area and time) can be attributed to a surface area where the emissions originate. Flux footprint models are commonly used to estimate this area of the emission origin. We used an updated version of the KL04−2D model that has been shown to perform best for airborne fluxes.[27] A graphical depiction of the footprints along our flight tracks is shown in Figure 1. This KL04−2D parameterization, available online,[46] was developed from a 1D backward Lagrangian stochastic particle dispersion model,[47] improved by Metzger et al.[48] to resolve the dispersion perpendicular to the main wind direction. The input parameters to the footprint model include the altitude of the measurements, standard deviation of horizontal and vertical wind speed, horizontal wind direction, boundary layer height, surface roughness length, and friction velocity. The obtained 90th percentile footprints—i.e., contouring the area where 90% of the measured emissions originate—were typically 3−6 km long and 2−6 km wide at their widest point and were then overlapped with census tracts to attribute the distribution of emissions to census tracts. Only census tracts with at least three flyovers, at least 10% of the respective footprint covering the respective census tract from a flyover, and the combined coverage of all flyovers being at least 100% were considered in our analysis.

For comparison of airborne HAP observations with the CARB emission inventory at the census tract level, we used the results of comparing our VOC measurements with the 2021 CARB inventory discussed in detail in Pfannerstill et al.[27] The measured−inventory differences at 4 × 4 km^2 grid cell level were matched to census tracts by calculating the weighted averages according to the fractional overlap of each census tract with each inventory grid cell.

2.5. Analysis Robustness and Uncertainties in Observations and Inequalities. The inequalities calculated in this study are not affected by the accuracy of emission values derived from airborne flux measurements. The accuracy (trueness) of these observations is on the order of ± 100%,[27] however, this uncertainty mainly stems from a scaling factor applied to all data to correct for vertical flux divergence, i.e., a meteorological phenomenon. Any correction to this error would be a uniform scaling factor applied to all data simultaneously, thus keeping relative inequalities the same. For a detailed description of these corrections, refer to the SI of Pfannerstill et al.[49] Likewise, the accuracy of mixing ratios

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(concentrations) is on the order of 10−50%. Since each HAP is always compared to itself in different locations in this study, this error also does not affect the inequalities calculated. Rather, any correction to it would be a scaling factor applied to all data points of the respective compound.

Measurements of HAPs and trace gases for sampled census tracts were averaged across the entire flight dataset (or population-weighted values calculated) to provide a single, representative data point within each analysis. The process of population weighting and averaging over all census tracts improves the precision of the reported results. The precision (i.e., random error) for each data point in our analysis was calculated following the approach of the standard error for unweighted averages—i.e., the 1 standard deviation of individual measurements of each bin (e.g., census tract or population group) were divided by the square root of the number of data points that were binned into each value (e.g., population-weighted concentrations). These random uncertainties are the standard mean errors and are represented by error bars in the x or y directions when visibly present in a figure. For mixing ratio (or emission) inequalities of demographic groups and communities, uncertainties were calculated for each HAP following the standard error for unweighted averages propagation.

Additionally, the US Census ACS comprises sampling and non-sampling errors,[50,51] which influence the accuracy and precision of calculated HAP inequalities. When random, these errors are also managed through population-weighting and averaging across many census tracts.

3. RESULTS AND DISCUSSION

3.1. Census Tract Differences in HAP Concentrations. Flight tracks for airborne eddy covariance measurements over the LA study area covered parts of Long Beach, Los Angeles City, Orange County, including Anaheim, and San Bernadino Valley (Figure S1). The study area spans neighborhoods that are home to some of LA's diverse populations, including Asians, Hispanics, and Latinos (hereafter, "Hispanics"), and census tracts with a range of MHI. A total of 1083 census tracts were sampled over all nine flights (Table S1), with coverage of major race-ethnicity groups—35% Hispanic, 30.6% non-Hispanic/Latino white (hereafter, "white"), 12% Asian, and 4.2% Black and African American—being representative compared to the LA region (see Text S2).[41] Given that the coverage of each demographic group by our flight tracks was not equal, we performed a sensitivity analysis (Text S2) to determine the number of airborne observations necessary to calculate robust population-weighted values for the sampled demographic groups. A minimum of ∼200 census tracts observed from all flights with at least 20% of a given demographic group in each tract was revealed to be necessary to calculate robust population-weighted values (Figure S2). Thus, to prevent bias in our results, we exclude Black and African Americans from our analysis, with only 66 observations of census tracts due to flight restrictions surrounding the predominantly Black neighborhoods around the LAX airport (Text S2 and Figure S1C).

We first evaluated the spatial distribution of HAPs in LA as a joint density function of census tract-level MHI and fractional race-ethnicity populations, separated into the lowest (0−25%), intermediate (>25−75%), and highest (>75%) percentiles of HAP concentrations. Figure 2 shows the concentration density distributions of benzene, naphthalene, p-chlorobenzotrifluoride (PCBTF), and NO_x for Hispanic populations, with an alternative representation provided in Figure S3. Lower HAP concentrations were associated with census tracts having fewer Hispanics and higher income residents, whereas areas with intermediate and higher concentrations of HAPs were more likely to be in tracts with lower household incomes and greater populations of Hispanics. Similar results were observed for all 17 measured HAPs and carbon monoxide (CO), as shown in Figure S4.

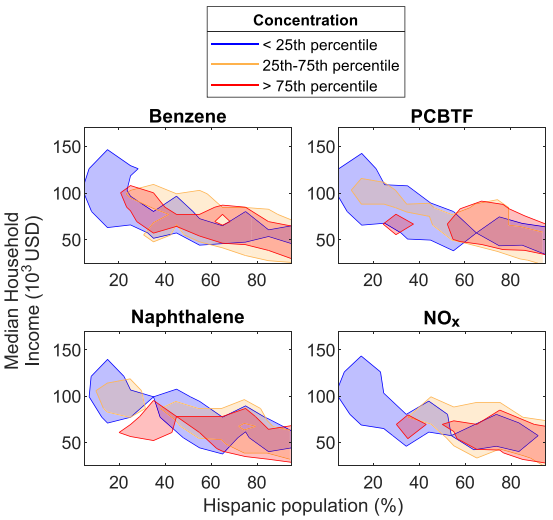

Figure 2. Contour plots surrounding the highest density of data points for the lowest 25%, intermediate 50%, and highest 25% of hazardous air pollutant concentrations plotted as a function of median household income and Hispanic population fractions per sampled census tract.

This trend is present but less apparent for Asians (Figure S5) due to closely clustered distributions of HAP concentrations that make it somewhat challenging to observe distinct census tract-level differences compared to Hispanic populations (Figure 2 and Figure S4). This is attributed to the spatial distribution of Asians in the study area (Figure S1A); unlike Hispanics, there were fewer census tracts with high Asian populations (>70%). Asians and Hispanics had isolation indexes[42] of 0.36 and 0.67, respectively, further reflecting that Asians are less isolated from their minority population in the LA study area, while Hispanics are more isolated and reside more closely with each other. This explains the less pronounced association observed in Figure S5 for increasing HAP concentrations and Asian populations compared to Hispanics (Figure 2 and Figure S4). Nonetheless, the concentration distribution of HAPs, NO_x, and CO, shown in Figure 2 and Figures S4 and S5, were consistent among low-income and non-white populations in LA and other large metropolitan areas.[11,16,40]

3.2. Inequalities in HAP Emissions and Concentrations. Since we performed spatially resolved measurements of not only concentrations but also emission fluxes, we were able to compare emission-based inequalities with those of concentrations. As emissions measurements in this study are not impacted by chemical transformations or atmospheric transport,[52] analyzing emission inequalities in comparison to concentration inequalities helps assess how localized emissions and meteorological conditions may shape HAP disparities, distinguishing source-driven from transport-driven impacts.

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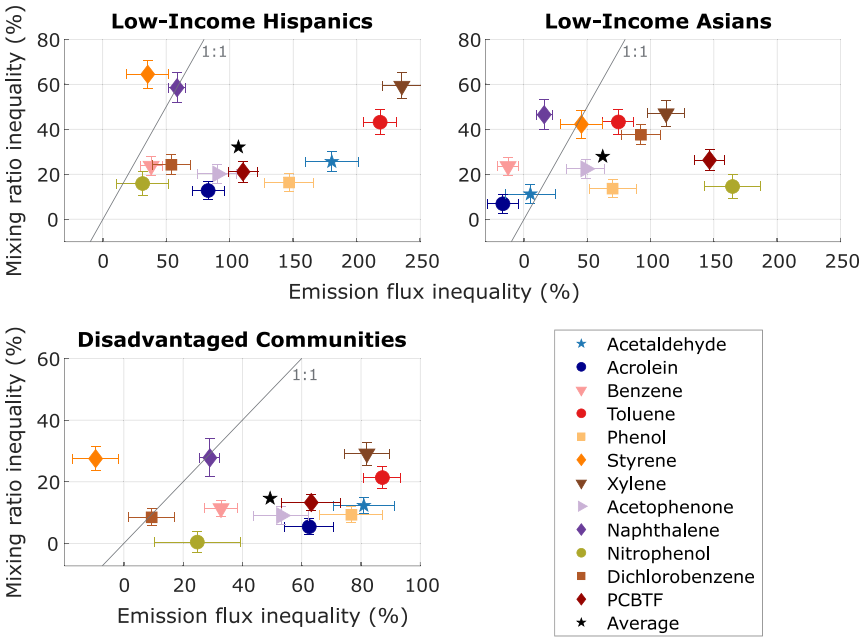

Figure 3. Inequality in fluxes (emissions) vs inequality in mixing ratios (concentrations) for hazardous air pollutants with a population-weighted emission above the 2σ method detection limit (see Data S1).[45] For low-income Hispanics (LIHs) and low-income Asians (LIAs), inequality is defined as the % difference in population-weighted values compared to that of high-income whites. Note the different *x*- and *y*-axis scale for Disadvantaged Communities (DACs). For DACs, inequality is the % difference in average concentration or emission values compared to those of non-DACs. Uncertainty (±) in HAP mixing ratios and emission fluxes are the standard mean errors (Table S3) of population-weighted values for demographic groups (LIAs and LIHs) and of the means for community (DACs) census tracts.

Population-weighted concentrations and emissions for Hispanic, Asian, and white populations were calculated for low- and high-income census tracts in the LA Basin (Section 2.2). Figure 3 (with values provided in Table S3) shows the relative inequalities in HAP concentrations and emissions calculated for low-income Hispanics (LIHs) and low-income Asians (LIAs) compared with high-income whites. On average (±SME), mixing ratio inequalities for LIAs and LIHs are 28 ± 4 and 32 ± 5%, respectively, while emission flux inequalities are 62 ± 17% for LIAs and 107 ± 21% for LIHs. Results show larger inequalities in emissions than concentrations for LIHs and LIAs, indicating that these population groups live in closer proximity to emission sources on average. We performed similar calculations solely based on income (Data S1 and Figures S6 and S7), and we found that LIHs and LIAs consistently experience higher HAP concentrations and inequalities than the broader low-income populations in LA, underscoring the needed context of considering race-ethnicity alongside income in our regional analysis (Text S1). We see similar results (Figure 3) for census tracts designated as Disadvantaged Communities (DACs) in California[43] within the study area (Figure S1E). DACs in the LA region are among the 10% of the most burdened communities in California and are the most heavily impacted by inequality in the distribution of pollution sources.[29] Average mixing ratio and emission flux inequalities for DACs are 15 ± 3 and 49 ± 9%, respectively, indicating that they overlap with LIA and LIH communities and reflect a share of nonwhite and economically disadvantaged populations.

We attribute the higher inequalities in HAP emissions, compared to concentrations (Figure 3), to atmospheric dispersion, chemical lifetime, and mixing as a result of dominant southwesterly winds during the study period. These winds are characteristic of mid-spring and summer months in Los Angeles.[53] Figure S8 depicts this effect more clearly for three example HAPs, benzene, acrolein, and PCBTF, where the spatial distribution of emissions is higher closer to the coast compared to higher concentrations situated farther inland in the sampled area. During the daytime in spring and summer months, southwesterly winds typically disperse pollutants more effectively (Figure S9A), resulting in an offset in HAP concentrations relative to emissions. This reduces the impacts of polluting sources partly for LIHs, LIAs, and DACs (Figure 3); however, since northerly and easterly winds make up more than 50% of the wind directions during winter months in LA,[53] it can be expected that inequality in concentrations will increase for LIHs, LIAs, and DACs under stagnant meteorological conditions (e.g., inversions; Figure S9B) characteristic of cooler periods in the region.[54]

While the number of flights in this study (Table S1) limits our ability to statistically infer associations between HAP inequalities and atmospheric conditions for different periods in LA, previous research has shown that slower winds reduce mixing for NO_x away from emitting sources and worsen inequalities.[28] In our study, the average (±standard deviation) daytime wind speed from all flights was 3.8 ± 1.3 m/s (Table S1), a range typical for summer days in LA.[53] In contrast, wind speeds in the region (Burbank, CA) averaged only 2.2 m/s during winter months in 2021.[53] These slower winds, combined with stronger inversions (Figure S9B), suggest greater accumulation of HAPs due to reduced atmospheric mixing in DACs and census tracts home to LIAs and LIHs. Thus, concentration inequalities for these populations are likely to be larger in winter than in summer.

To demonstrate the influence of meteorological conditions on pollutant concentrations, we draw on ground-based HAP concentration data in the LA region (discussed in Text S3; see Table S2 and Figure S10).[23] In predominantly Hispanic

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communities, surface HAP concentrations increase markedly from summer to winter (e.g., benzene: 0.148 ± 0.029 ppb in summer vs 0.74 ± 0.226 ppb in winter in Compton, CA) (Table S2 and Figure S10), as reduced atmospheric mixing during colder months traps emissions locally. In contrast, in predominantly white communities, where emissions are lower, concentrations remain relatively stable across seasons (e.g., benzene: 0.241 ± 0.031 ppb in summer vs 0.251 ± 0.046 ppb in winter in Burbank, CA). Thus, the difference between HAP concentrations measured in the predominantly Hispanic and white community increases from −39% in summer to 195% in winter for benzene—comparable to our average observed inequality jumping from 32% in concentrations to 107% in emissions for LIHs (Figure 3). This means that any results from concentration measurements in Los Angeles at daytime in summer—typical time for research campaigns and measurements—will underestimate annual average inequalities. This underscores the importance of our emission flux measurements, which, assuming relatively constant emissions throughout the year, capture the high inequality potential for LIHs, LIAs, and DACs in the region, independently of meteorology-induced chemical transport.[52] From a policy perspective, our findings emphasize the need to account for meteorological conditions to fully understand the extent of HAP concentration inequalities. Targeted emission reductions in low-income and minority communities would reduce the emission inequalities we reported and, consequently, the concentration inequalities, especially when they are at their worst—in winter.

Considering the adverse health outcomes associated with these relatively long-lived HAPs,[5−7] higher concentrations raise relevant concerns from a public health perspective. For instance, benzene, toluene, xylene, and styrene are HAPs primarily associated with industrial activity (e.g., petrochemical and manufacturing) and mobile sources—as evaporative emissions and products of incomplete combustion.[55] Our analysis shows that for all observations of these HAPs among LIHs, LIAs, and DACs, relative concentration inequalities were up to 60% (Figure 3 and Table S3), with absolute inequalities ranging from 0.01 to 0.09 ppb (Figure S11). We also evaluated weekday−weekend differences given their correlation with traffic-related pollutants such as NO_x and CO[56] and discuss these results in the SI (see Text S4 and Figures S12 and S13). Results from weekday−weekend differences in HAP inequalities point to contributions not only from traffic but also from non-vehicular sources, particularly volatile chemical products (VCPs).[18] Among measured urban VOCs in the LA study region, purely solvent-sourced PCBTF was shown to be among the top 10 VOCs both for emissions mass flux and SOA formation potential.[27] PCBTF is a carcinogenic VOC-exempt solvent used in the production of VCPs such as inks, paints, and cleaners.[57] Notably, higher emissions and concentrations of PCBTF in the study area (Figure S8E,F) coincided with census tracts having large Hispanic populations (Figure S1B). These findings reflect ongoing concerns raised in predominantly Hispanic communities of East, South, and Southeast LA on increasing emissions and associated health concerns from solvents like PCBTF,[58−60] prevalent from numerous local sources such as autobody shops, dry cleaners, manufacturing facilities, and related industries.[61] Thus, replacing PCBTF and similar solvent-containing VCPs with less harmful options should be considered toward addressing concerns and reducing inequalities in these communities.

In this study, many of the measured HAPs[31] lack regulatory inhalation risk data, making it difficult to accurately assess potential cancer or health risks for the sampled census tracts. Presently, only six out of the 17 measured HAPs—acetaldehyde, acrylonitrile, benzene, naphthalene, dichlorobenzene, and PCBTF—possess a screening target risk level corresponding to a one-in-a-million [10^{-6}] chance for cancer.[62] Concentrations of acetaldehyde, benzene, and PCBTF we observed in the LA Basin exceed their respective carcinogenic screening levels[62] for all groups (Data S1). This points to the impact of pervasive traffic in LA, influencing concentrations of benzene and acetaldehyde (also a byproduct of secondary formation), as well as increasing solvent emissions from the growing use of PCBTF.[27] Furthermore, it is important to consider the likely cumulative impacts[63] on LA's low-income residents and communities of color, as these findings underscore the potential compounding effects of HAP emissions, further exacerbating health disparities.[64]

3.3. Identifying Sources of Inequalities in HAP Emissions. Los Angeles presents a complex array of air pollution sources, posing challenges in precisely attributing HAP emissions to individual sources. Understanding the source of emissions is necessary for targeted regulation and mitigation efforts, protecting public health, and addressing environmental justice concerns. The emission ratio of toluene and benzene (T/B) can be useful in distinguishing likely sources of HAP emissions,[65−70] due to their frequent co-emission among urban ambient VOCs and other HAPs. Additionally, our emission flux data provide surface emission values for toluene and benzene, unimpacted by atmospheric oxidation.[45] Thus, for our analysis, we examined candidate sources from major known emission sources in LA,[18] which are distinguishable by their T/B ratios. These include diesel traffic and oil and gas production (T/B < 1.5),[65,66] gasoline vehicle emissions (T/B ≈ 2),[67,68] industrial processes (2.4 < T/B < 3.2),[69] and solvent use sources (3.2 < T/B < 6).[70]

Figure 4 illustrates T/B ratios for the LA study region binned by fractional Hispanic and Asian populations as well as DACs and non-DACs. In areas with low Hispanic populations (<25%), the median T/B ratio reflects the source profile of gasoline exhaust (Figure 4), likely stemming from widespread traffic and freeway emissions prevalent in LA. However, as Hispanic populations increase (>25%), there is a shift toward higher T/B ratios, indicating that industrial and solvent use sources become important as well. Interestingly, census tracts with the highest Hispanic populations (>75%) exhibit the highest median T/B ratios. This suggests that in addition to traffic, industrial processes and solvent use have become significant contributors to HAP emissions and inequality in densely populated Hispanic census tracts. This trend persists when comparing DACs and non-DACs and is consistent with previous assessments on the disproportionate siting of pollution sources and industries in DACs across LA.[29] Oil and gas activities in LA have also been areas of concern and air pollution exposure for Black and Hispanic populations as well as DACs residing near production sites.[71] T/B emission ratios associated with oil and gas operations are typically lower than other HAP sources (<2) given high-sourced benzene levels.[66] As a result, our observations, which are characterized by high T/B ratios, do not indicate a strong influence of oil and gas sources on the inequalities impacting Hispanics and DACs in our dataset.

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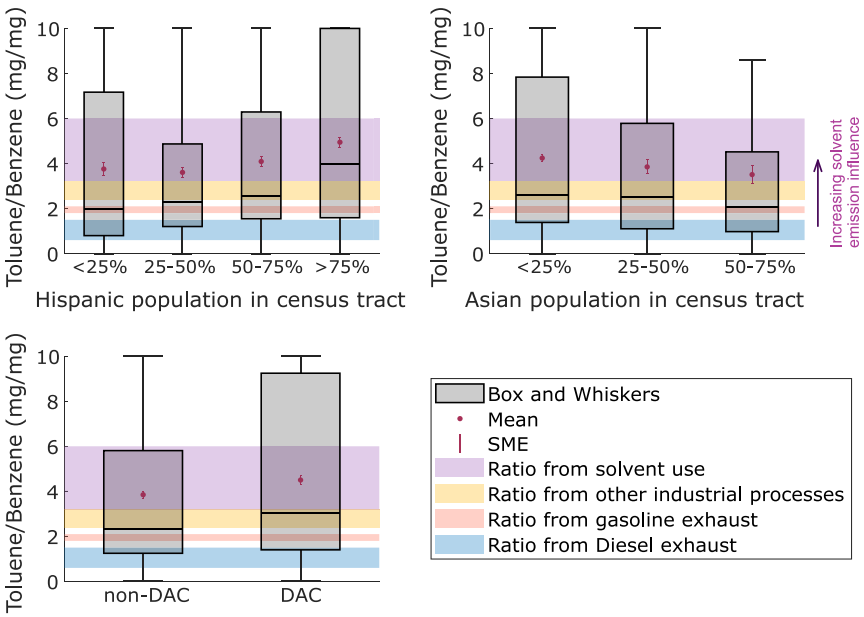

Figure 4. Toluene-to-benzene emission ratios as a function of binned population fraction in respective census tracts for Hispanics and Asians, and binned by Disadvantaged and non-Disadvantaged Communities. Boxes represent the 25th−75th percentiles, whiskers represent the 5th−95th percentiles, points represent the means, and horizontal lines represent the medians. Uncertainty (±) in the means are the standard mean errors (SME) (Table S4). Shaded colored areas show the range of toluene-to-benzene emissions from four different emission sources.[65−70] Note that the ratios are based on surface emissions already corrected for oxidative loss.[27] The >75% bin for Asian population contained only 10 data points and was therefore omitted as not statistically significant.

For Asian populations, emission sources contrast with Hispanics and DACs. In census tracts with lower Asian populations (<50%), industrial processes are the main HAP emissions source, reflected by median T/B ratios (Figure 4). As Asian populations increase in sampled census tracts, the T/B ratio decreases, indicating a shift to traffic-related emissions. This trend is likely influenced by Asians residing in census tracts (Figure S1A) with higher proportions of other racial-ethnic groups so that a high fraction of Asians likely corresponds to a lower fraction of Hispanics (Figure S1B).[41] As a result, higher Asian populations in LA appear to be situated in census tracts where typical traffic and freeway emissions are the predominant HAP sources. This agrees with observed weekday−weekend differences discussed in the SI, comparing traffic and potential solvent impacts for LIAs and LIHs (Text S4 and Figures S12 and S13), and aligns with US-wide $PM_{2.5}$ exposure disparities showing that Asians are most heavily impacted by pollution surfacing from light-duty gas vehicles in contrast to industrial sources for Hispanic communities.[38]

We also see a widespread of T/B ratios with mean values consistently higher than the medians (Figure 4). This suggests that solvent use is impacting the high end of toluene emissions, which spatially vary in the LA region. Although benzene and toluene are both constituents in vehicle fuel, toluene is used more abundantly in various industries compared to benzene, which is more tightly regulated due to its carcinogenicity.[72] The wide spatial distribution and high mean-to-median values of T/B ratios across sampled tracts indicate growing non-traffic (solvent) sources for toluene amid decreasing benzene emissions. This is consistent with an average T/B ratio of 4.1 for the entire LA study area[27] compared to previous measurements of 2.9 in 2010.[20] Likewise, recent studies report that non-vehicular industrial sources, mainly attributed to VCPs, now contribute as much as 50% of anthropogenic VOC emissions in LA, previously dominated by mobile sources.[18,26,73] Furthermore, these results appear to primarily stem from business-as-usual emission sources in LA, as HAP observations during the study period rule out potential influences from biomass burning events (Text S5).

Overall, our observations of source-specific emissions underscore the need for enhanced regulation of solvent sources, particularly VCPs, which our data show to be the main drivers of inequality in HAP emissions. It is essential that the use of hazardous solvents in industries, including commercial establishments like autobody shops, be carefully scrutinized, particularly in minority and Disadvantaged Communities where these sources are concentrated. Special attention should be given to unregulated toxic air pollutants such as PCBTF.

3.4. Census Tract-Level Differences in Measured and Inventory HAP Emissions. Emission inventories from regulatory agencies such as the California Air Resources Board (CARB) are essential for chemical transport models used to estimate air quality and inform policymaking in large metropolitan regions such as LA. We were able to compare CARB inventory estimates, comprising anthropogenic (point and mobile) and biogenic sources, with our airborne eddy covariance measurements in LA. The spatial resolution of our directly measured emissions enabled matching with census tracts, which allowed the validation of the emissions inventory at the same scale. A comprehensive evaluation of measured and inventory VOC emissions in LA can be found elsewhere.[27] Here, we focus on measured−inventory emission differences for only six of the 17 studied HAPs available from the CARB inventory, grouped by fractional census tract Hispanic populations (Figure 5). Notably, PCBTF[57] is absent from the CARB inventory, underscoring additional needs in regulatory accounting of pertinent industry compounds and solvents contributing to toxic VOC emissions.[18,73]

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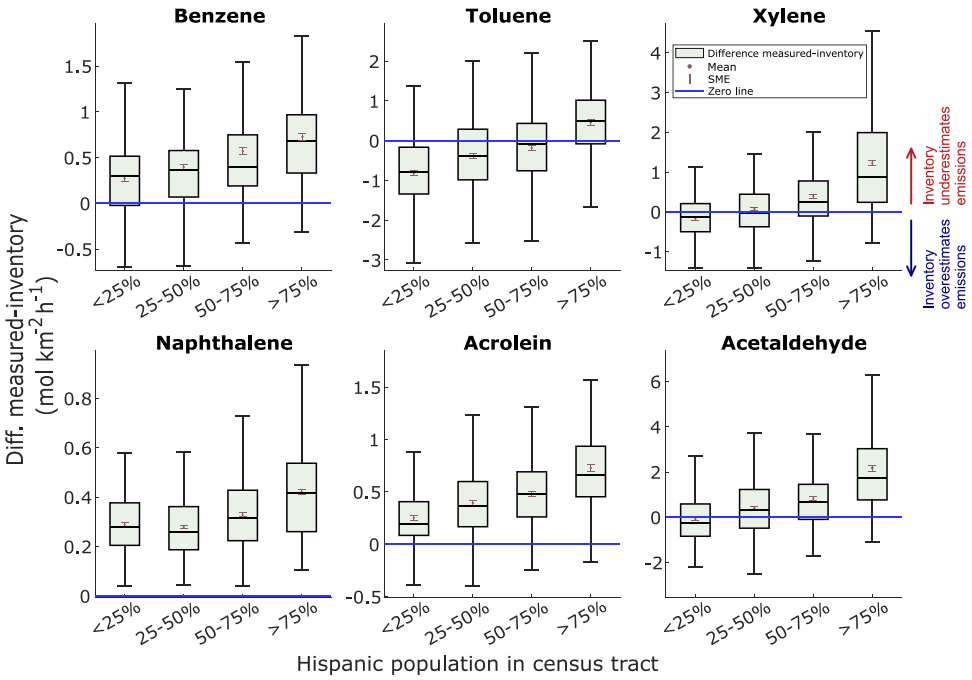

Figure 5. Differences in measured and inventory HAP emissions binned by fractional Hispanic populations across sampled census tracts. A positive difference above the zero line signifies underestimation of the observed emissions by the CARB inventory. The zero line is shown in blue, boxes represent the 25th−75th percentiles, whiskers represent the 5th−95th percentiles, points represent the means, and horizontal lines represent the medians. Uncertainty (±) in the means are the standard mean errors (SME) (Table S5). A simple one-way ANOVA, not including the uncertainty of HAP measurements, indicated a statistical significance of $p < 0.001$ for measured-inventory differences across binned Hispanic population fractions for the presented HAPs.

In areas with higher Hispanic populations, the CARB inventory underpredicts HAP emissions to a greater extent (Figure 5), scaling with fractional census tract populations, where measured fluxes are also higher (Figure S14). Conversely, lower median differences in measured and inventory HAP emissions are observed in census tracts with low Hispanic populations and for non-DACs (Figure S15), indicating better performance of the inventory in these areas. These results have implications for largely populated Hispanic communities and DACs in LA. For instance, while toluene and xylene emissions show relatively better agreement with the CARB inventory (Figure 5), benzene exhibits much higher measured inventory differences. Given benzene's well-documented carcinogenicity,[72] this underestimation of emissions indicates likely underprediction of the associated health risks in these communities. Additionally, uncovering the demographic bias in a widely used regulatory emission inventory, which underpins air quality predictions and policy in California, highlights the need for more timely and accurate regulatory accounting of HAPs necessary for protecting the health of populations with environmental justice concerns.

3.5. Implications. This study explored spatially resolved concentrations and emissions of HAPs and other gaseous air pollutants in the LA Basin, and contextualized observations with census tract-level information. Unlike previous literature on environmental equity and air pollution disparities, we performed direct emission measurements covering a whole megacity at high spatial resolution (~2 km). This approach enabled matching of emissions and concentrations of rarely observed air toxics with specific census tracts without relying on modeling. These measurements also allowed for the quantification of inequalities in where emissions occur, as well as the identification of sources of the pollutants being emitted.

Study findings show higher population-weighted concentrations of the 17 measured HAPs, as well as NO_x and CO, in census tracts with lower household incomes and higher Hispanic populations (Figure 2 and Figure S4). Low-income Hispanics and Asians were more burdened by localized emission sources, with average HAP emissions 107 ± 21 and 62 ± 17% higher, respectively, compared to high-income whites (Figure 3). These emissions show the high inequality potential for LIHs and LIAs under less favorable meteorological conditions throughout the year. Summertime in LA reflects when inequalities in HAP concentrations are expected to be at their lowest, as a constant amount of emitted pollutants become more diluted due to increased atmospheric mixing and dispersion compared to winter conditions (e.g., inversions; see Figure S9). Nonetheless, HAP observations indicated that industry- and solvent-related impacts diverged among Hispanics and Asians (Figure 4). Specifically, census tracts with larger Hispanic populations (>50%) had higher median toluene-to-benzene ratios (>3) indicative of industrial processes and solvent use emission sources. Furthermore, PCBTF, a carcinogenic and purely solvent-sourced VOC,[57] exhibited higher emissions over sampled census tracts densely populated by Hispanics compared to those with high Asian or white populations (Figure S8E,F).

We also compared emission measurements to the performance of VOC inventories in overburdened communities for the first time. Results reflected lags in the regulatory accounting of key HAPs like PCBTF, raising concerns for environmental justice and public health. Emissions of several HAPs were also notably underestimated, with the severity increasing with the

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fraction of Hispanic populations in census tracts (Figure 5 and Figure S14).

There are some limitations to the analysis presented in this study, particularly related to potential biases from capturing only a subset of the entire census tract population in the LA Basin through our sample flights. This limitation is particularly relevant for Black and African American populations, as this group was not sampled sufficiently due to flight route restrictions (Text S2 and Figure S1C). Given that Black and African Americans have historically been among the most disadvantaged in the United States[74] and are likewise disproportionately impacted by air pollution in LA,[8,14,15,75,76] it is reasonable to assume that the HAP inequalities observed in our study would be similar, if not more pronounced, in these populations.

Additionally, compared to numerous studies on air pollution disparities in the LA region,[8−11] our HAP measurements cannot directly link concentration inequalities to exposure implications, which is often a key factor in public health analyses. However, our approach focused on emissions measurements provides valuable insights into actual emission rates. Additionally, since the airborne eddy covariance method is independent of chemical transport,[52] the results can be integrated into inventories and models to simulate various weather scenarios and infer exposure risks—processes that typically depend on such inventories. This approach offers a robust foundation for understanding emission inequalities, which are the underlying source of exposure in the context of public health.

Although emission measurements (the eddy covariance method) are not influenced by meteorology-induced chemical transport, meteorology itself can impact emissions. Thus, evaporative emissions like solvents, asphalt, or evaporation of gasoline can be higher on warm summer days, such as during our study period (see Figure S16).[45] This means that the VCP emissions reported here may be on the higher end of the annual average. However, the impact of temperature on these emissions is smaller than wintertime inversion effects that cause much larger inequalities to occur in winter—as shown in year-round ground-based measurements in the LA region (Table S2 and Figure S10).

Finally, the PTR-ToF-MS method employed in this study is limited by its mass range (~30 to 390 amu) and blind to low-mass alkanes (e.g., oil and gas sources), inorganic HAPs (e.g., heavy metals), or low-polarity halogenated compounds. However, this method is particularly sensitive to aromatic compounds, which are important HAPs given their toxicity, as well as oxygenated VOCs that are often not observed at continuous observation stations equipped with gas chromatography techniques. A comparison of the organic gaseous compounds in the California air toxics inventory for the South Coast AQMD[77] with the species we observed, weighted by emitted mass and divided by chronic inhalation reference concentrations,[62] showed that we observed 74% of the inventoried toxicity (Figure S17). Additionally, the non-targeted nature of the PTR-ToF-MS was a key advantage, allowing for the detection of compounds not previously considered as relevant air toxics, such as PCBTF, in this study.

Overall, efforts to reduce hazardous air pollutant inequalities in DACs and communities with large Hispanic populations must address the sources of increasing VCP emissions in Los Angeles. Additionally, obtaining accurate data on the emissions and concentrations of HAPs must be prioritized by regulators for better inventory estimates that drive decision-making, aimed at mitigating disparities and reducing overall health risks for impacted populations. Predominantly Hispanic communities, particularly East, South, and Southeast LA, have been designated as AB 617 communities[58−60] focused on achieving air pollution emissions reductions from various sources, including truck traffic, freeways, and solvent-use industries. Concerns in these communities[57,59] have led to a proposed revision of Rule 1171, which regulates solvents, by the South Coast AQMD to prohibit the use of PCBTF.[78] If approved, this regulatory action will likely have direct impacts across LA and yield air quality benefits, particularly for large Hispanic communities in the region. Thus, the findings in this study may be well positioned to infer census tract-level HAP inequalities, thereby providing additional support for impacted communities in continuing the push for city-wide decision-making to address disparate impacts. The spatial distribution and inequality of HAPs in Los Angeles underscore the importance of addressing air quality issues in the region to achieve meaningful and equitable outcomes.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.5c00808.

Supporting Texts S1−S5; Figures S1−S17; Tables S1−S5; flight tracks and meteorological conditions; sensitivity analysis of sampled census tracts; HAP concentrations, emissions, and inequalities for demographic groups and communities; and maps, absolute fluxes, and measured-inventory differences of HAP emissions (PDF)

(Supporting Data S1) Summary tables of HAP mixing ratios and emission fluxes for study groups (XLSX)

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#J.O. and E.Y.P contributed equally; J.O. wrote the manuscript and supported data analysis; E.Y.P. performed measurements together with C.A., conducted the data analysis, and supported manuscript development; S.E.P. provided expertise and supported data analysis; C.E.I. provided expertise and supported manuscript development; and A.H.G. supervised the study and oversaw data analysis.

Notes

The authors declare no competing financial interest.

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