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Observing Nitrogen Dioxide Air Pollution Inequality Using High-Spatial-Resolution Remote Sensing Measurements in Houston, Texas

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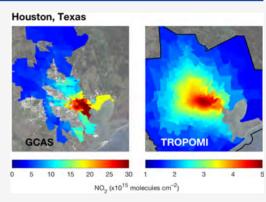
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ABSTRACT: Houston, Texas is a major U.S. urban and industrial area where poor air quality is unevenly distributed and a disproportionate share is located in low-income, non-white, and Hispanic neighborhoods. We have traditionally lacked city-wide observations to fully describe these spatial heterogeneities in Houston and in cities globally, especially for reactive gases like nitrogen dioxide (NO₂). Here, we analyze novel high-spatial-resolution (250 m \times 500 m) NO₂ vertical columns measured by the NASA GCAS airborne spectrometer as part of the September-2013 NASA DISCOVER-AQ mission and discuss differences in population-weighted NO₂ at the census-tract level. Based on the average of 35 repeated flight circuits, we find 37 \pm 6% higher NO₂ for non-whites and Hispanics living in low-income tracts (LIN) compared to whites living in highincome tracts (HIW) and report NO₂ disparities separately by race ethnicity (11–32%) and poverty status (15–28%). We observe substantial time-of-day and day-to-day variability in LIN-HIW NO₂ differences (and in other metrics)



driven by the greater prevalence of NO_x ($\equiv NO + NO_2$) emission sources in low-income, non-white, and Hispanic neighborhoods. We evaluate measurements from the recently launched satellite sensor TROPOMI (3.5 km × 7 km at nadir), averaged to 0.01° × 0.01° using physics-based oversampling, and demonstrate that TROPOMI resolves similar relative, but not absolute, tract-level differences compared to GCAS. We utilize the high-resolution FIVE and NEI NO_x inventories, plus one year of TROPOMI weekday—weekend variability, to attribute tract-level NO_2 disparities to industrial sources and heavy-duty diesel trucking. We show that GCAS and TROPOMI spatial patterns correspond to the surface patterns measured using aircraft profiling and surface monitors. We discuss opportunities for satellite remote sensing to inform decision making in cities generally.

■ INTRODUCTION

Houston, Texas is a large, socio-demographically diverse U.S. city that is also a global center for petrochemical manufacturing. Houston experiences among the worst air quality in the United States^{1,2} with documented evidence that local air pollution disproportionately burdens the city's low-income residents and communities of color,³⁻⁵ leading to demonstrated differences in health and life expectancy as a result.^{6,7} Similar racial, ethnicity, and income-based inequalities have also been observed in other major U.S. cities.⁸⁻¹³ Our ability to describe intra-urban pollutant variability has been limited by the lack of spatially continuous, temporally resolved measurements that capture gradients between neighborhoods. In the case of reactive gases with short atmospheric lifetimes, such as nitrogen dioxide (NO₂), intra-urban spatiotemporal variability cannot be directly observed by traditional monitoring approaches, impeding efforts to address air quality disparities through policy.

 NO_2 plays a critical role in surface air quality, and knowledge of NO_2 spatiotemporal variability is fundamental to air pollution and public health decision making. NO_2 is a key control over the atmospheric oxidation capacity, a precursor to ground-level ozone and particulate matter (PM), and a regulated criteria pollutant under the Clean Air Act. In U.S. cities, NO_2 emissions are typically dominated by mobile sources and fossil-fuel-based electricity generation; in Houston, petrochemical refineries and industrial activities are also large emitters. $^{15-17}$ NO_2 is a robust indicator of combustion emissions generally 10 and a common surrogate

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for traffic-pollutant mixtures, especially toxic diesel exhaust. ¹⁸ Epidemiological studies have linked ambient NO₂ concentrations to adverse health outcomes, ^{19–22} and residential proximity to roadways (determined using geospatial and/or NO₂ datasets) has been associated with reduced lung function and asthma, ^{23–25} cardiac and pulmonary mortality, and preeclampsia and preterm birth. ^{26–28}

NO2 concentrations change rapidly in the near-field of sources with downwind distance-decay gradients, largely controlled by rates of dispersion, ranging from <0.5 km in unstable to 1-2 km in stable atmospheres. 29,30 Widely used monitoring tools such as regulatory surface networks and satellite instruments have historically been unable to resolve such NO2 gradients. Across the United States, there are over 400 in situ NO₂ monitoring stations (typically measuring NO₂*, representing NO₂ plus a positive measurement bias), yet fewer than 1/3 of U.S. urban areas are equipped with even one analyzer.³¹ In cities with monitors, monitoring stations are spatially sparse, and, while urban NO2 is well correlated with traffic, U.S. Environmental Protection Agency (EPA) guidance generally recommends siting monitors away from roadways. 31,32 At the same time, satellite remote sensing has contributed substantially to our understanding of inter-urban NO₂ distributions, ^{31,33–35} providing spatially continuous maps of NO2 columns across cities, but mostly lacked the resolution required to capture intra-urban variability. The Ozone Monitoring Instrument (OMI) with a nadir footprint of 13 km × 24 km has previously been shown to detect near-surface NO₂ variability within large cities, ^{31,36} including individual slant-column observations when operated in the "super-zoom" mode $(13 \text{ km} \times 3 \text{ km})$. Fine spatial scale land-use regression (LUR) models, which predict NO2 concentrations as a function of the geographic location of emitters and other land cover elements, have added spatial detail to coarse NO2 datasets³⁸⁻⁴⁰ and allowed the creation of NO₂ maps at resolutions of 100 m × 100 m, fine enough to resolve distancedecay gradients away from sources. However, these models require substantial a priori geospatial knowledge that may not be available and rely on temporal averages of pollutant data of at least weeks (surface monitors) to months (satellites). As a result, LUR models typically do not describe NO2 temporal variability, hindering source identification and discovery using time-varying emission patterns and leading to biases in acute exposure epidemiological studies related to confounding temporal trends and meteorology¹⁸ with analyses based on emission inventories and pollutant proxies, e.g., neighborhood distance to roadways, facing similar limitations.

Here, we evaluate the extent that higher-resolution NO2 remote sensing can resolve intra-urban spatiotemporal variability relevant to NO2 inequality in Houston, Texas. First, we describe an observationally based analysis using novel fine-spatial-resolution (250 m × 500 m), sub-orbital remote sensing measurements from NASA GCAS (GEOstationary Coastal and Air Pollution Events (GEO-CAPE) Airborne Simulator)^{41–43} collected as part of the September 2013 DISCOVER-AQ study (Deriving Information on Surface Conditions from COlumn and VERtically Resolved Observations Relevant to Air Quality). We quantify neighborhood-level (census-tract) differences in population-weighted tropospheric NO2 vertical columns, with GCAS columns resolving censustract variability, and discuss atmospheric controls over the NO2 spatiotemporal distribution for various socio-demographic groups, specifically race-ethnicity and income. Second, we

test whether the recently launched satellite-based TROPOMI sensor, currently producing the highest spatial resolution NO₂ satellite measurements over North America, precisely and/or accurately captures the same NO2 column differences detected by GCAS. We utilize in situ NO2 vertical profiles collected onboard the NASA P-3B during DISCOVER-AQ and data from the routine surface monitoring network to demonstrate that columns and surface measurements represent similar NO₂ spatial patterns. We evaluate major source contributions to census-tract-level NO2 disparities, comparing weekday-weekend column differences in the first full year of TROPOMI data (June 2018 to May 2019) to estimates derived from emissions in the high-resolution Fuel-based Inventory of Vehicle Emissions (FIVE) and National Emission Inventory (NEI). Finally, we discuss potential applications and limitations of next-generation satellite observations to inform, evaluate, enforce, and motivate decision making on air pollution inequality in U.S. cities.

■ MATERIALS AND METHODS

Houston, Texas. Houston-The Woodlands-Sugar Land, referred to in this paper as either Houston or the Houston Metropolitan Area (HMA), is the largest metropolitan statistical area in Texas and among the largest in the U.S. with 6.1 million residents in 2013 and 6.8 million in 2018. Houston is a growing city that is racially, ethnically, and economically diverse and home to one of the largest Hispanic populations in the U.S. Houston is also the location of $\sim 1/4$ of all U.S. chemical refineries⁴⁵ and the Houston Ship Channel (HSC), a busy waterway where numerous major industrial facilities are located, extending from the Gulf of Mexico, through Galveston Bay, and along the Buffalo Bayou river between Baytown and Downtown Houston (Figure S1).⁴⁶ The unique combination of urban transportation, petrochemical emissions, and the prevalent land-sea breeze⁴⁷ contribute to the HMA's poor air quality, including high levels of ozone⁴⁸ and hazardous air pollutants. 49 Past research has shown that local air pollution is not uniformly distributed across Houston but instead concentrated in neighborhoods with larger Hispanic populations, lower rates of educational attainment, and higher rates of poverty.^{3,4} Primarily non-white, Hispanic, and lower-income neighborhoods have experienced greater cancer risks, increased chronic and acute air pollutant exposure, and lower overall physical well-being, especially in communities adjacent to facilities on the HSC where lowincome households and people of color are statistically overrepresented.6,7

GCAS. The Geostationary Coastal and Air Pollution Events (GEO-CAPE) airborne simulator (GCAS) was developed as a technology demonstration instrument in support of the Geostationary Coastal and Air Pollution Events (GEO-CAPE) decadal survey mission.⁵⁰ GCAS makes hyperspectral nadir-viewing measurements of backscattered solar radiation in two channels at wavelengths of 300-490 nm (air quality species) and 480-900 nm (ocean color). Each channel uses a two-dimensional charge-coupled device (CCD) array detector for mapping with one CCD dimension capturing absorption spectra and the other providing cross-track spatial coverage over an ~45° field of view. GCAS NO2 column retrievals consisted of a two-step approach similar to algorithms used for other major satellite instruments: GOME, SCIAMACHY, OMI, TROPOMI, and the forthcoming TEMPO. 41,42 First, NO₂ slant column densities were derived by direct spectral

fitting of radiances using measured nadir spectra and an averaged unpolluted reference spectrum (over the Gulf of Mexico). Second, slant columns were converted to vertical columns using an air mass factor (AMF), which was a function of the observing geometry, surface reflectance, ozone profile, and trace gas profile shape. AMFs were calculated for each scene with scattering weights derived from a radiative transfer model over 56 vertical layers, 45 of which were generated from the Community Multi-scale Air Quality (CMAQ). CMAQ (version 5.0.2) was run at a spatial resolution of $4 \text{ km} \times 4 \text{ km}$, temporal resolution of 20 min, and vertical resolution ranging from 22 m (surface) to ~200 m (2 km) to ~650 m (aircraft altitude) with offline meteorology driven by the Advanced Research Weather and Forecasting (WRF-ARW) model⁵¹ using the Meteorology-Chemistry Interface Processor (MCIP).52 Details on the CMAQ and WRF modeling can be found in the work of Loughner and Follette-Cook, 53 and further details on other aspects of the GCAS instrument and retrieval can be found in the work of Nowlan et al. 41,42 GCAS column uncertainties were estimated to range between 20 and 50% and 18 and 30% over moderately $(0.5-1 \times 10^{16})$ molecules cm⁻²) and heavily polluted (>2 \times 10¹⁶ molecules cm⁻²) areas, respectively. 41 Column uncertainties were driven by uncertainties in the AMF and challenges associated with representing species exhibiting high spatiotemporal variability as means. Nowlan et al. 41 reported a small bias (+2%) in the GCAS columns from the use of CMAQ-modeled NO₂ profiles. GCAS observed individual spectra with native spatial resolution on the order of tens of square meters, but spectra were spatially averaged to enhance their signal-to-noise ratio. At a 9 km flight altitude, GCAS produced NO₂ vertical columns at 250 m (across track) × 500 m (along track) with the along-track coverage generated by the host aircraft.

During the Houston DISCOVER-AQ deployment, GCAS flew onboard the NASA B-200 and conducted a total of 21 air quality flights in the morning (8 am-12 pm, LT) and afternoon (1 pm to 5 pm, LT) over 11 days. In this analysis, we focused on measurements of cloud-free pixels from nine weekdays that included both morning and afternoon research flights, typically with two circuits during each flight: September 4, 6, 11–13, 18, and 24–26, 2013. Data from the second circuit of the morning flight of September 12 were omitted because of heavy clouds.

TROPOMI. The TROPOspheric Monitoring Instrument (TROPOMI) is one of the newest space-based NO₂ sensors 54,55 and the single payload onboard the sunsynchronous Copernicus Sentinel-5 Precursor (S5P) satellite. TROPOMI measures in the ultraviolet and visible (270-500 nm), near-infrared (675-775 nm), and shortwave infrared (2305-2385 nm) spectral regions to quantify a range of atmospheric trace gases. NO₂ is retrieved by fitting the 405-465 nm band using an updated OMI DOMINO algorithm and based on work from the QA4ECV project. 56-60 At nadir, NO₂ is retrieved at a spatial resolution of 3.5 km \times 7 km for the time period of this study. 54,55 Precision of individual tropospheric NO2 columns over urban/polluted scenes is on the order of 30-60%⁶¹ and dominated by uncertainties in the AMF. Key inputs to the AMF are: clouds; the NO2 profile shape, generated using $1^{\circ} \times 1^{\circ}$ TM5-MP model output; ⁶² and the surface albedo, from a $0.5^{\circ} \times 0.5^{\circ}$ monthly OMI climatology.⁶³ We used Level 2 NO₂ tropospheric columns, quality descriptor: qa value of >0.75 as recommended by the Product User Manual.64

TROPOMI maps at $0.01^{\circ} \times 0.01^{\circ}$ were produced using physics-based oversampling. In brief, the approach represents observations on the ground as sensitivity distributions, rather than as points or polygons. For image grating spectrometers like TROPOMI, generalized two-dimensional super Gaussian functions have been shown to best characterize the sensitivity distributions. For details on the oversampling method, see Sun et al. 65

Surface NO₂* and Winds. Hourly ground-based nitrogen dioxide (NO2*), wind speed, and wind direction measurements were downloaded via the Texas Commission on Environmental Quality Data Report query tool (https:// www17.tceq.texas.gov/tamis/index.cfm). NO₂* was measured using chemiluminescence coupled to a heated molybdenum converter that includes a positive interference from nitric acid and organic nitrates, 66 which largely affects absolute rather than relative NO₂* mixing ratios. We use the nomenclature NO₂* rather than NO₂ in acknowledgment of this interference. For September 2013, we analyzed wind data from 16 monitoring stations in the HMA with simultaneous NO2* and wind observations. To observe interannual trends, we compared daytime (10 am to 4 pm LT) annual averaged data for June 2013 to May 2014 and June 2018 to May 2019 using NO₂* measurements from the 15 stations operating in both time periods. To compare surface and TROPOMI column observations, we included mean midday (12-3 pm LT) measurements from all 17 monitors in the HMA, excluding two designated near-roadway sites⁶⁸ (see Figure S14 caption for station information).

Demographic Data and Boundaries. Household income statistics from the 2010 American Community Survey: five-year dataset and race-ethnicity population tables from the 2010 U.S. Census were downloaded from the IPUMS National Historical Geographic Information System. ⁶⁹ Year 2013 census tract polygons were downloaded as TIGER/Line shape files from the data.gov library: https://www.census.gov/cgi-bin/geo/shapefiles/index.php.

P-3B Dataset and Convective Boundary Layer (CBL) Height Determination. As part of DISCOVER-AQ, NO₂ and NO were measured onboard the NASA P-3B at a 1 s time resolution by the NCAR chemiluminescence system, detecting NO directly and NO₂ following photolysis into NO by a blue light converter. The instrument was calibrated frequently in flight, and uncertainties were 0.02 ppb in precision (at 1 s averaging) and $\pm 10\%$ accuracy for NO₂. H₂O_(v) was measured by the NASA open-path diode laser hygrometer (DLH) and reported at 1 s time resolution with overall uncertainties of 5%. Static air temperature data were collected by a Rosemount model 102 sensor with a precision of 0.006 °C and accuracy of ± 0.2 °C.

For each P-3B profile, we identified the CBL height as the altitude of the strongest coincident gradients in potential temperature (θ) and water vapor mixing ratio $(H_2O_{(v)})^{,70}$ which were also near the top of the region of constant $H_2O_{(v)}$ mixing ratios extending from the surface. To reduce spurious layer determination from instrument noise, we first averaged measurements into 10 m altitude bins and calculated 5-point running mean lapse rates and $H_2O_{(v)}$ gradients. Individual CBL heights for each profile are given in Table S2. Vertical profiles of NO2, lapse rate, θ , and $H_2O_{(v)}$ with their corresponding CBL heights over Moody Tower, Channelview, Deer Park, West Houston, Conroe, and Manvel Croix are provided in the Supporting Information.

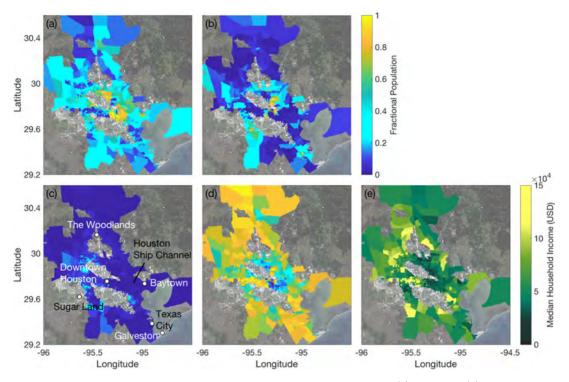


Figure 1. Fractional census-tract-level race-ethnicity demographics along the GCAS flight tract: (a) Hispanics, (b) Black and African Americans, (c) Asians, and (d) non-Hispanic whites. Panel (c) includes labels for various locations within the HMA. (e) Fractional median annual household income in USD. While the aircraft flew a repeated circuit, there were slight variations in some circuits between flights; this exact circuit is from the morning of September 4. Background map data: Landsat 8 composite over January 2017 to June 2018.

FIVE and NEI Inventories. The Fuel-based Inventory of Vehicle Emissions (FIVE) is a spatially (500 m \times 500 m) and temporally (monthly) resolved inventory of mobile source emissions (on-road + off-road engines).⁷² FIVE provided on-road emissions derived from publicly available fuel sales reports, road-level traffic counts, and time-resolved weigh-inmotion traffic counts.⁷³ Fuel use uncertainties are based on differences between fuel sale reports and truck travel ($\pm 13\%$ in Texas) and traffic count site selection and sample size ($\pm 10\%$ for major roads and freeways in large urban areas). Emissions uncertainties were derived from regression analysis of nearroad infrared remote sensing and tunnel studies: ±16% and ±15% for light-duty gasoline vehicles and heavy-duty diesel trucks, respectively.⁷² We focused on annual averages of the on-road component, which was comprised of light-duty gasoline vehicles and heavy-duty trucks with fuels sales and emission factors updated to 2018.

The National Emissions Inventory (NEI) provides a comprehensive and detailed estimate of NO_2 emissions from stationary sources, including industrial facilities, power plants, airports, and commercial facilities. We used estimates from the 2014 NEI Version 1, encompassing reports from state, local, and tribal air agencies and the EPA programs: Toxic Release Inventory, Acid Rain Program, and Maximum Achievable Control Technology standards development. Emission uncertainties in power plants were $\pm 25\%$, and errors in industrial facilities and other less-characterized stationary sources were assumed to be $\pm 50\%$.

Population-Weighted NO₂ and Census-Tract-Level NO₂ Differences. GCAS and TROPOMI NO₂ columns were averaged within census tract polygons and tagged with geographic identifiers. Population-weighted NO₂ columns were calculated as equal to the product of the tract-unit NO₂

column $(NO_{2,j})$ and demographic group population (p_j) (by race ethnicity or poverty classification) in the ith tract, summed over all census tracts with NO_2 data (n) and divided by the summation of the group population (p_j) (eq 1). Errors were defined as standard mean errors with N representing the number of census tracts with both NO_2 observations and residents of a given demographic group.

population-weighted NO_{2,j} =
$$\sum_{i=1}^{n} NO_{2,i} p_{i,j} / \sum_{i=1}^{n} p_{i,j}$$
 (1)

We discussed absolute and relative NO₂ inequality in terms of the absolute and percent difference in population-weighted NO₂ columns between two demographic groups. Raceethnicity demographics were defined with the following U.S. Census codes: Black and African Americans (JMJE004), excluding individuals identifying as Hispanic or Latino; Asians (JMJE006), excluding those identifying as Hispanic or Latino; Hispanics (JMJE012), including all races reporting as Hispanic; and non-Hispanic whites (JMJE003). GCAS sampling was statistically representative of race-ethnicity demographics in the HMA (GCAS, HMA): Black and African American (17%, 12%), Asian (5%, 4%), Hispanic (36%, 37%), and non-Hispanic white (42%, 47%). Census-tract-level race, ethnicity, and income variability along the GCAS flight track is shown in Figure 1. We calculated NO2 for non-whites and Hispanics living in low-income tracts, denoted as LIN, by first applying an income threshold of census-tract median household incomes of <35,000 USD and then computing population-weighted NO2 columns in the low-income tracts for Black and African Americans, Asians, and Hispanics. Likewise, we calculated NO₂ columns for whites living in highincome tracts (HIW), applying the income threshold of

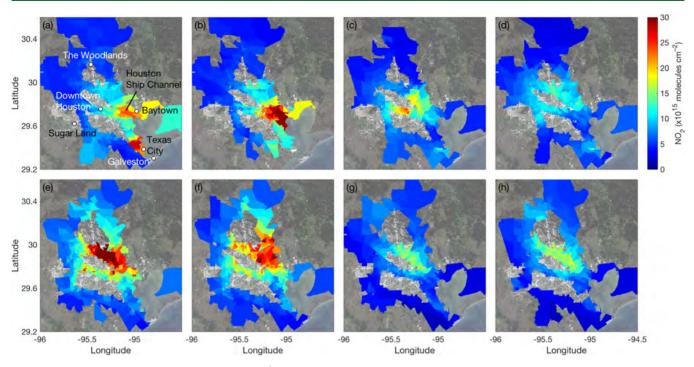


Figure 2. GCAS NO₂ column densities (molecules cm⁻²) averaged to census tracts on two sample days, (a–d) September 4 (top row) and (e–h) 26 (bottom row), during eight flight circuits in the early morning (8 am–10 am LT; panels a and e), late morning (10 am–12 pm LT; panels b and f), early afternoon (1 pm–3 pm LT; panels c and g), and late afternoon (3 pm–5 pm LT; panels d and h). Background map data: Landsat 8 composite, January 2017–June 2018.

Table 1. Census-Tract-Averaged Population-Weighted NO₂ Columns (molecules cm⁻²) for Weekdays (Tuesday–Friday) during Morning and Afternoon GCAS Flights (Average of the Two Circuits) with Variability Reported as Two Standard Deviations of Individual Circuit Population-Weighted NO₂ Means for Different Socio-Demographic Categories; Annual (June 2018 to May 2019) TROPOMI Observations Sub-sampled along the GCAS Flight Track (September 4, Morning) on Weekdays and for the HMA on Weekdays and Weekends (Saturday–Sunday); September 2018 Weekday TROPOMI Observations, both along the GCAS Flight Track (September 4, Morning) and for the Full HMA^a

	population-weighted census-tract-averaged NO $_2$ (× 10^{15} molecules cm $^{-2}$)											
		GCAS weekd	ay flights		TROPOMI							
	means		variability (2σ)		annual (along GCAS)	annual (HMA)		September (along GCAS)	September (HMA)			
group	morning	afternoon	morning	afternoon	weekdays	weekdays	weekends	weekdays	weekdays			
LIN	13.9 ± 0.5	9.0 ± 0.3	15.3	4.3	3.7 ± 0.1	3.3 ± 0.1	2.6 ± 0.1	4.7 ± 0.1	4.0 ± 0.1			
HIW	9.1 ± 0.4	6.3 ± 0.3	4.2	2.4	2.7 ± 0.1	2.2 ± 0.1	1.9 ± 0.1	3.1 ± 0.1	2.9 ± 0.1			
below-poverty	13.0 ± 0.4	8.4 ± 0.3	13.7	4.1	3.5 ± 0.1	3.1 ± 0.1	2.1 ± 0.1	4.4 ± 0.1	3.7 ± 0.1			
near-poverty	11.3 ± 0.2	7.5 ± 0.1	9.8	3.3	3.2 ± 0.1	2.9 ± 0.1	2.0 ± 0.1	4.0 ± 0.1	3.4 ± 0.1			
above-poverty	9.5 ± 0.3	6.5 ± 0.2	5.1	2.5	2.8 ± 0.1	2.6 ± 0.1	1.9 ± 0.1	3.4 ± 0.1	2.9 ± 0.1			
Hispanic	12.8 ± 0.4	8.4 ± 0.2	13.3	3.9	3.5 ± 0.1	3.3 ± 0.1	2.2 ± 0.1	4.5 ± 0.1	4.0 ± 0.1			
Black/African American	11.4 ± 0.4	7.2 ± 0.2	8.3	3.3	3.2 ± 0.1	3.0 ± 0.1	2.1 ± 0.1	3.9 ± 0.1	3.4 ± 0.1			
Asian	10.4 ± 0.3	6.7 ± 0.2	3.3	2.4	3.0 ± 0.1	2.8 ± 0.1	2.0 ± 0.1	3.6 ± 0.1	3.3 ± 0.1			
white	8.8 ± 0.3	6.2 ± 0.2	4.9	2.7	2.7 ± 0.1	2.3 ± 0.1	1.7 ± 0.1	3.1 ± 0.1	2.5 ± 0.1			
^a GCAS and TROPOMI	uncertainties	are standar	d mean er	rors round	ed up to at least	one decima	l position.					

census-tract median household incomes of >80,000 USD then computing population-weighted NO_2 columns in high-income tracts for non-Hispanic whites. For reference, lower (upper) annual household-income quintiles were 34,588 (84,905) USD along the GCAS flight path and 33,860 (79,332) USD across the nine counties of the HMA. Along the GCAS track, 13% of the population met the criteria for LIN (16% for HIW), residing in 18% of census tracts (23% for HIW); in the HMA, 16% of the population met the criteria for LIN (13% for HIW), residing in 25% percent of tracks (17% for HIW). Poverty status was categorized following the U.S. Census Bureau

definition using the household income-to-poverty ratio. Households were classified as being below the poverty line if their income was less than the poverty threshold in the U.S. Federal Poverty Guidelines, which scales with the number of people per household. We defined census tracts as below poverty if >20% of households in the tract were at or below an income-to-poverty ratio of 1. The corresponding number of households was used in eq 1. Households with an income-to-poverty ratio greater than 1 in the remaining census tracts comprise the above-poverty population. Near-poverty pop-

ulations encompassed households in all tracts with an incometo-poverty ratio of 1-1.24.

■ RESULTS AND DISCUSSION

GCAS, Census-Tract NO₂ Differences, and Temporal Variability. We focused on GCAS measurements from 35 flight circuits, collected on 9 weekdays (Tuesday-Friday) with sampling conducted in both the morning (8 am-12 pm local time, LT) and afternoon (1 pm-5 pm LT) (Figures S2-S10). Observations from two example days are shown in Figure 2 with GCAS vertical columns averaged to the underlying census tracts. To investigate differences in neighborhood-level NO2, we calculated tract-level race-ethnicity population-weighted mean NO₂ columns for census tracts along the GCAS flight path for each circuit and compared absolute and relative column differences (Tables 1 and 2). Population-weighted NO2 columns for Hispanic, Black and African American, and Asian residents across all tracts sampled by GCAS were higher than for non-Hispanic whites by $32 \pm 11\%$, $19 \pm 7\%$, and $11 \pm$ 5%, respectively (Table 1). In census tracts defined as below or near the poverty line, NO₂ columns were on average $28 \pm 11\%$ and $15 \pm 8\%$ higher than those above the poverty line. For non-whites and Hispanics in low-income tracts, populationweighted NO₂ columns were $37 \pm 6\%$ higher than for HIWs. Correspondingly, predominantly Hispanic and Black/African American neighborhoods and lower-income neighborhoods of all races and ethnicities were more often located in central Houston and in closer proximity to the HSC (Figure 1). Because we utilized population-weighted columns, our results do not simply reflect urban-rural gradients. In the U.S. Census-designated "urbanized areas" only (Figure 3b), population-weighted NO2 columns for Hispanic, Black and African American, and Asian residents were $21 \pm 5\%$, $13 \pm 6\%$, and $3 \pm 6\%$ higher than for whites, respectively. In census tracts below and near the poverty line, population-weighted NO_2 columns were 24 \pm 6% and 14 \pm 5% higher than for those above the poverty line, and, for LINs, populationweighted NO_2 columns were $31 \pm 8\%$ higher than for HIWs. Nowlan et al.41 reported that GCAS was found to underestimate NO2 columns compared to DISCOVER-AQ P-3B measurements for higher column densities with GCAS lower by 10% and overestimate columns near background concentrations with GCAS higher by $\sim 1.6 \times 10^{15}$ molecules cm⁻². As a result, census-tract-level NO2 differences reported here are likely biased low and NO₂ inequalities are underestimated.

While population-weighted NO2 columns were always higher for LINs than for HIWs, a wider range of NO2 levels were observed in predominantly non-white, Hispanic, and lowincome tracts (Table 2). Variability, defined as two standard deviations of mean population-weighted NO2 for all 35 flight circuits, was a factor of 2.5 greater for LINs than HIWs. On September 25, meteorological conditions contributed to latemorning NO₂ columns and LIN-HIW differences (84 \pm 9%) that were uniquely high (Table 2 and Figure S9); however, even after removing all September 25 data, populationweighted NO2 columns were still 1.8 times more variable for LINs than HIWs. In the afternoon on all days, absolute NO₂ column densities were lower for both LINs and HIWs than in the morning. Afternoon mean surface winds were $8 \pm 1 \text{ m s}^{-1}$ and usually from the southeast (onshore flow); by comparison, morning surface winds were 6 ± 2 m s⁻¹ and, while typically from the east/east northeast, more varied in direction (Table S1). The observed variability indicated greater NO_x emission

Table 2. GCAS Census-Tract-Averaged Population-Weighted NO₂ Columns (molecules cm⁻²) for Each Flight Circuit between LINs and HIWs and Their Percent

			difference (%)	28 ± 4	21 ± 5	23 ± 6	45 ± 6	41 ± 3	20 ± 3	15 ± 3	32 ± 4	44 ± 3	30 ± 6
afternoon third circuit fourth circuit	ighted NO_2 cules cm^{-2})	HIW	7.4 ± 0.4	5.9 ± 0.3	6.3 ± 0.4	6.5 ± 0.3	5.7 ± 0.2	5.7 ± 0.2	5.7 ± 0.2	10.0 ± 0.5	7.0 ± 0.2	6.7 ± 0.3	
	population-weighted NO ₂ $(\times 10^{15} \text{ molecules cm}^{-2})$	TIN	9.9 ± 0.2	7.2 ± 0.4	8.0 ± 0.4	10.2 ± 0.5	8.6 ± 0.4	7.0 ± 0.3	6.6 ± 0.2	13.8 ± 0.5	12.0 ± 0.4	9.1 ± 0.4	
		difference (%)	44 ± 7	36 ± 7	33 ± 9	47 ± 7	45 ± 7	19 ± 5	17 ± 5	35 ± 6	55 ± 6	39 + 6	
	third circuit	third circuit ighted NO ₂ cules cm ⁻²)	HIW	7.8 ± 0.5	6.0 ± 0.3	5.7 ± 0.4	4.8 ± 0.2	5.3 ± 0.2	5.4 ± 0.2	4.8 ± 0.2	8.9 ± 0.5	5.8 ± 0.2	6.0 + 0.3
	second circuit	population-weighted NO (X 10 ¹⁵ molecules cm ⁻²)	LIN	12.3 ± 0.5	8.6 ± 0.4	7.9 ± 0.4	7.7 ± 0.3	8.3 ± 0.4	6.6 ± 0.2	5.7 ± 0.2	12.6 ± 0.5	10.3 ± 0.4	8.9 + 0.4
			difference (%)	39 ± 7	24 ± 6	20 ± 9		14 ± 7	13 ± 7	2 ± 6	84 ± 9	34 ± 6	40 + 8
		second circuit ighted NO ₂ cules cm ⁻²)	HIW	8.3 ± 0.5	6.0 ± 0.3	8.3 ± 0.6		10.2 ± 0.5	7.6 ± 0.4	6.2 ± 0.2	15.4 ± 1.3	12.4 ± 0.5	9.3 + 0.5
morning	population-weighted NO (× 10 ¹⁵ molecules cm ⁻² .	LIN	12.4 ± 0.5	7.6 ± 0.3	10.2 ± 0.5		11.8 ± 0.5	8.7 ± 0.4	6.3 ± 0.3	37.8 ± 1.9	17.3 ± 0.7	14.0 + 0.6	
			difference (%)	28 ± 5	28 ± 6	42 ± 7	25 ± 6	41 ± 7	17 ± 7	4 ± 6	58 ± 6	61 ± 7	34 + 8
	first circuit	first circuit sighted NO ₂ cules cm ⁻²)	HIW	7.5 ± 0.3	8.9 ± 0.4	8.1 ± 0.4	8.7 ± 0.4	9.0 ± 0.5	8.0 ± 0.3	8.1 ± 0.4	13.1 ± 0.7	11.2 ± 0.4	9.2 + 0.4
		population-we	population-weighted NO (× 10 ¹⁵ molecules cm ⁻²	LIN	10.0 ± 0.4	11.8 ± 0.4	12.4 ± 0.5	11.1 ± 0.4	13.7 ± 0.6	9.4 ± 0.5	8.5 ± 0.3	23.9 ± 0.9	21.2 ± 1.0
			September	4	9	11	12	13	18	24	25	26	mean

^aFor individual circuits, uncertainties are standard mean errors of population-weighted NO₂ across all tracts within the flight path. The second morning pass on September 12 was omitted because most of the circuit was not completed and there were too many clouds

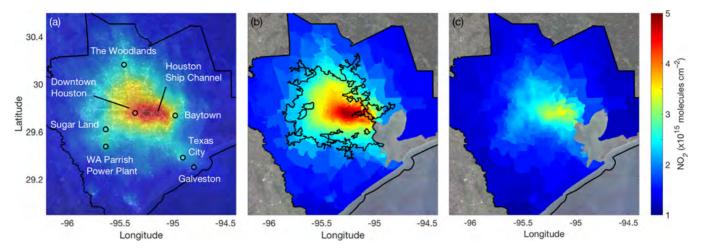


Figure 3. (a) Weekday (Tuesday–Friday) TROPOMI NO_2 column densities (molecules cm⁻²) for June 2018–May 2019 at $0.01^{\circ} \times 0.01^{\circ}$ over the greater Houston area and (b) averaged within census tracts. (c) Weekend (Saturday–Sunday) TROPOMI NO_2 columns averaged within census tracts for the HMA. A 40% transparency is applied to the TROPOMI data in panel (a) to see the underlying land cover. The inner black outline in panel (b) depicts the census "urbanized area" and the outer black depicts the HMA. Background map data: Landsat 8 composite, January 2017 to June 2018.

source density in proximity to non-whites living in low-income neighborhoods as the highest concentrations should co-locate with sources for reactive gases. We test this conclusion against emission inventory data (below) as high-income tracts were also 60% larger than low-income tracts, leading to greater spatial averaging.

Evaluating TROPOMI Observations. GCAS operates as a satellite analog in NASA airborne missions and its high-spatial-resolution observations now provide data for TROPOMI validation. Launched in October 2017, TROPOMI has been shown to generate detailed NO₂ column maps, revealing hotspots undetectable by past space-based sensors due to its order of magnitude-improved spatial resolution. A6,77–79 Here, we use the high-spatial-resolution, but limited duration, GCAS dataset to evaluate TROPOMI-derived annual differences in census tract-scale NO₂ columns, both along the GCAS flight track and across the HMA. The comparison provides an evaluation of the suitability of TROPOMI observations to resolve key horizontal NO₂ gradients for assessing air pollution disparities.

In Figure 3a,b, TROPOMI observations are weekdays (Tuesday–Friday) over June 2018 to May 2019 with data collection occurring at $\sim\!1:30$ pm LT. Mondays were excluded as they were considered transition days. TROPOMI measured highly localized NO2 column enhancements over the HSC, Texas City Galveston Bay Refinery (the second largest refinery in the U.S.) and the W.A. Parrish Generating Station (a 3.65 GW dual-fired power plant that includes the largest coal-fired plant in Texas at 2.7 GW). To produce TROPOMI maps at $0.01^{\circ}\times0.01^{\circ}$ ($\sim\!1$ km \times 1 km over Houston), we employed a physics-based oversampling of daily images. The annual-average weekday NO2 pattern was spatially similar to GCAS measurements on days with slower surface winds, onshore afternoon air flows, and higher-than-average NO2, in particular, September 4, 25, and 26.

Population-weighted annual weekday TROPOMI columns were 31 \pm 3% higher for LINs than for HIWs when sampled along the GCAS flight track (Figure S11). Across the full HMA (Figure 3b and Table 1), weekday TROPOMI observations indicated 34 \pm 2% higher population-weighted NO₂ columns for LINs than for HIWs. In Houston's "urbanized areas" only

(Figure 3b), which included 79% of tracts in the HMA, population-weighted LIN NO_2 columns were $27 \pm 3\%$ higher than for HIWs. Finally, in the month of September only (2018), LIN population-weighted weekday TROPOMI NO_2 columns were $42 \pm 3\%$ greater than for HIWs. Results from both annual and September TROPOMI weekday means agreed with the corresponding GCAS LIN-HIW differences measured during the third circuit (1–3 pm) of $39 \pm 6\%$ to within uncertainties. Between 2013 and 2018, surface-level NO_2^* mixing ratios averaged across the HMA decreased by $6 \pm 5\%$ (1.0 ± 7 ppb) with no major changes in the NO_2^* spatial distribution and lower absolute and relative changes recorded at monitors nearest to the HSC compared to sites in suburban HMA (Figure S12).

In addition to somewhat lower relative LIN-HIW differences, the annually averaged TROPOMI results yielded lower absolute weekday LIN-HIW differences (9.19 × 10¹⁴ molecules cm⁻²) compared to those from GCAS (2.6×10^{15} molecules cm⁻²). Temporally averaging TROPOMI observations over the course of a year would have reduced the impact of individual high pollution episodes that exacerbated inequality, such as on September 25, and led to smaller LIN-HIW differences compared to September means; declines in overall NO₂ between 2013 and 2018 may have played a role as well. Lower absolute column densities were also consistent with TROPOMI's coarser spatial resolution and documented TROPOMI biases, with columns biased low under polluted conditions (high NO₂) and biased high at low NO₂ levels, which have been attributed to the lower resolution surface albedo characterization (0.5° OMI LER) and the coarse resolution of the TM5-MP a priori profiles used in the standard product.^{77,80} While a similar analysis for TROPOMI has not yet been published, McLinden et al.81 found that a custom high-resolution OMI retrieval reduced the AMF over NO₂ plumes compared to the DOMINOv2 (similar to the retrieval used here) where the vertical column is equal to the slant column/AMF. The ratio of the custom retrieval AMF to the DOMINOv2 AMF was maximal at 1.9 over the NOx source but decreased to 1 on a spatial scale relevant to an urban-suburban gradient, and, as a result, TROPOMI observations likely underestimate the severity of NO₂

disparities. That being said, we have found that annually averaged TROPOMI maps do capture a key portion of LIN-HIW differences (and inequality by other metrics) compared to those from GCAS despite the effects of temporal averaging and biases.

TROPOMI and afternoon GCAS measurements were compared as joint probability density functions of household income and fractional census tract race-ethnicity sorted into the lowest (0-20% of the NO_2 column distribution), intermediate (40-60%), and highest- NO_2 (80-100%) quintiles; Figure 4 shows the median contours of each density

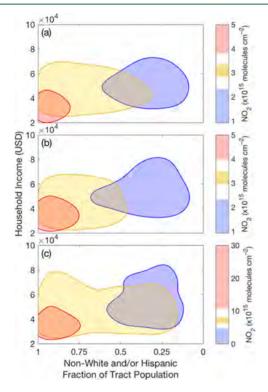


Figure 4. HMA population as a function of census tract annual household income (USD) and non-white/Hispanic or non-Hispanic white fractions separated by a census-tract-averaged NO $_2$ column quintile: high NO $_2$ (80–100% highest column densities) (red), mid-quintile NO $_2$ (40–60%) (yellow), and low NO $_2$ (0–20%) (blue). (a, b) Annual (June 2018 to May 2019) weekday (Tuesday–Friday) TROPOMI observations across the HMA (a) and along the GCAS flight track (b). (c) Composite distribution of all 35 GCAS circuits. Color bars represent vertical column densities (molecules cm $^{-2}$) corresponding to the NO $_2$ quintiles.

function. The results showed that neighborhood-level differences were not driven by outliers but represented broader patterns in the NO₂ distribution with neighborhood demographics. TROPOMI and GCAS described similar patterns: low NO₂ census tracts were more likely to be white and of higher income, and intermediate and high NO₂ tracts were more likely to be non-white, Hispanic, and of low income. Figure 4 also illustrates that TROPOMI and GCAS represented the NO₂ column density distributions as normal or log-normal, respectively (represented in the color scales), which was consistent with the coarser spatial resolution of the TROPOMI observations and their reported low bias at high NO₂. In addition, while the high (red) and low (blue) quintiles were comparable between datasets, there was more variation in

the mid (yellow) quintile, reflective of these differences in the NO₂ distribution.

Finally, census tracts are aspatial administrative units optimally sized around 4000 people that represent fine spatial scales in cities. In Houston's "urbanized areas" (Figure S15), census tracts were on average $4.7 \pm 6.5 \text{ km}^2$ ($\sim 2 \text{ km} \times 2 \text{ km}$ if square) and the smallest 20% of tracts were on average 1.2 \pm 0.3 km². Because the smallest tracts are the size of our oversampled TROPOMI product resolution, finer-scale analyses are likely limited. On average, the even smaller administrative unit of the U.S. Census block group, optimally sized at \sim 1500 people, was 2.1 \pm 5.5 km² in the "urbanized" areas" and just $0.3 \pm 0.1 \text{ km}^2$ in the smallest 20% of tracts with the latter approaching the limit of the GCAS spatial resolution. At the block-group level, we computed LIN-HIW NO₂ differences of 22 \pm 5% for TROPOMI and 30 \pm 4% for GCAS (all circuits), which were 35% and 17% lower than computed at the tract level, respectively, suggesting even the GCAS resolution was too coarse to fully resolve block-groupscale disparities. While finer-scale data hypothetically reveal greater NO2 inhomogeneity and inequality, higher-resolution NO₂ remote sensing observations potentially underestimate the impacts of NO_x emissions. Because NO_x is primarily emitted as NO and then converted to NO2 in the presence of ozone, NO2 remote sensing misses the portion of NO2 temporarily stored as NO as the system reaches steady state. This is important for a city-wide NO2 column comparison in differently sized tracts, especially if using NO2 as a surrogate for other co-emitted species, as sources are more prevalent in low-income neighborhoods, non-white and Hispanic residents are statistically overrepresented in tracts with higher population densities, and ozone concentrations are spatially

NO₂ Column-Surface Relationships. Satellite and suborbital remote sensing instruments observe NO2 columns and precisely capture surface neighborhood-scale differences when NO2 vertical distributions do not co-vary with census-tract demographics. Daytime continental NO2 mixing ratios are typically highest within the convective boundary layer (CBL), the thin layer of air in contact with the Earth's surface. While long-lived species are generally well-mixed in the CBL, NO2 can exhibit steeper vertical gradients, as chemical loss and turbulent mixing timescales are similar. 82 To investigate the extent that NO2 columns represented surface-level patterns in tract-scale NO2 inequality, not the surface concentrations themselves, which is a key factor in their application to air pollution decision making, we analyzed spatiotemporal variability in in situ NO2 vertical profiles and TROPOMI columns to NO₂* surface measurements.

As part of DISCOVER-AQ, the NASA P-3B aircraft profiled the lower troposphere, spanning altitudes of 3–0.3 km at locations corresponding to the GCAS flight path (Figure S16). In their detailed evaluation of the GCAS retrieval during DISCOVER-AQ, Nowlan et al. showed that observations from GCAS and aircraft profiling were well correlated with an overall r^2 of 0.89. We focus on 144 profiles representing tracts of varied population demographics in downtown (Moody Tower), the HSC (Channelview and Deer Park), and suburban Houston (Conroe, West Houston, and Manvel Croix), collected on eight weekdays in the morning (8:30–10:30 am, LT), at midday (11:30 am–1 pm, LT), and in the afternoon (1:30–3:30, LT) (Figure S13). Profiling at Deer Park, Conroe, and West Houston included missed approaches

over lightly trafficked air strips, facilitating sampling below altitudes of 0.1 km. All profiles are provided in the Supporting Information.

CBL heights were on average 0.8 ± 0.2 km $(\pm 1\sigma)$ in the morning, 1.5 \pm 0.5 km at midday, and 1.7 \pm 0.6 km in the afternoon. CBL heights were not statistically different at any profile location, and temporal variability was generally well correlated across sites (Table S2). In the morning, the slope of the correlation between the 3 km column and within-CBL column (Table S4 and Figure S17) was 0.80 ± 0.18 ($r^2 =$ 0.99), and, once the CBL was fully developed (midday and afternoon), the slope was 0.98 ± 0.15 ($r^2 = 0.99$). The slope of correlation between the column within the CBL and below 500 m AGL was 0.83 ± 0.19 ($r^2 = 0.95$) in the morning. Once the CBL was developed, the slope decreased to 0.27 \pm 0.05 (r^2 = 0.80), consistent with atmospheric conditions in which timescales of turbulent mixing and NO2 chemical loss are competitive. The high correlation coefficients indicated that location-dependent differences in the NO2 vertical distribution were small and demographic-based NO2 comparisons would not substantially differ if derived from surface measurements instead of columns.

We then compared tract-averaged TROPOMI columns and daytime (12–3 pm LT) NO_2^* surface mixing ratios measured at monitors across the HMA (Figure 5). We linearly correlated

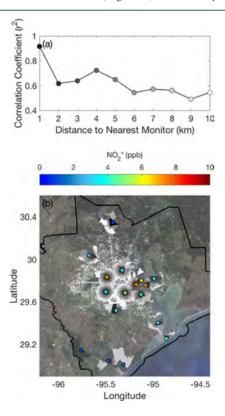


Figure 5. (a) Correlation coefficient between tract-averaged NO_2 measured at surface monitors and their overhead columns as a function of distance away from the monitor with respect to the census tract center point. NO_2^* observations are daytime (12–3 pm LT) averages and both datasets include weekdays (Tuesday–Friday) over June 2018 to May 2019. (b) Census tracts coloration indicates the distance (km) between the nearest surface NO_2 monitoring station and the census tract center point for tracts within a 10 km radius. Background map data: Landsat 8 composite over January 2017 to June 2018.

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annual (June 2018 to May 2019) weekday (Tuesday-Friday) mean observations as a function of the distance between tract center points and the nearest monitors. Surface NO2* and directly overhead columns (tracts within 1 km of monitors) were strongly correlated ($r^2 = 0.92$). However, r^2 decreased with increasing distance from the nearest monitor, falling to 0.63-0.75 when tracts were 2-5 km from the local monitor and 0.50-0.58 at distances of 5-10 km. Column-surface correlations have also been shown to improve with monitor density elsewhere using simulated TROPOMI datasets³⁶ and observations from the more coarsely resolved satellite OMI sensor.³¹ While an evaluation of the daily TROPOMI observations is the subject of a future manuscript, we investigated the temporal correlation between individual daytime (12-3 pm LT) mean surface NO₂* mixing ratios and overhead (within 1 km of a monitoring station) annual weekday TROPOMI NO2 columns (Figure 3b). The mean correlation coefficient, including days when at least 75% of monitoring stations were operational, was 0.66 ± 0.17 (see Figure S14 for histograms of the individual values) with no significant seasonal variation observed. While further analysis would have been required to infer surface concentrations, our results showed that TROPOMI captured typical NO₂ surface patterns, and, therefore, surface-level NO2 inequality, but the use of annual average TROPOMI observations obscured dayto-day variability in the surface NO2 distribution. The steep decline in r^2 at 1–2 km corresponded to previously observed NO₂ distance-decay gradients, ^{29,30} demonstrating the limits of the routine network to detect representative NO₂ levels for the majority of local residents, an advantage of TROPOMI, as just 3.4% of census tracts in the HMA (based on tract center points) were located within 2 km of a NO2 monitor.

NO_x Source Contributions to Census-Tract NO₂ **Differences.** To investigate the greater temporal variability in the GCAS results and observationally evaluate the contribution of heavy-duty diesel trucking emissions to inequality, we analyzed LIN-HIW differences reported in the FIVE and stationary source NEI using the same population weighting applied to the NO2 column observations. Total population-weighted emission densities (metric tons NO2-eq day⁻¹ km⁻²) (composed of on-road diesel and gasolinepowered vehicles, industrial and petrochemical facilities, and electricity generation) were 5 times higher for LINs than for HIWs. Gasoline-powered and heavy-duty diesel vehicle (HDDV) emission densities were 4 and 4.2 times greater for LINs than for HIWs, respectively. The largest disparities were found for industrial sources with 10 times greater emission densities for LINs than for HIWs. This unequal distribution of emission sources was consistent with our observation or more variable NO₂ concentrations and our inference of greater NO_x emissions in proximity to non-white, Hispanic, and low-income households.

Satellite remote sensing has the advantage of capturing temporal variability useful for interpreting the impact of NO_x sources, in particular, NO_2 differences on weekdays and weekends that are driven largely by HDDV traffic patterns. The HDDVs represent just 3–6% of the number vehicles in the U.S. vehicle fleet; however, because diesel engines produce \sim 7 times more NO_x per kilogram of fuel burned than gasoline, they contribute a substantial portion of NO_x emissions in many U.S cities. HDDVs have been shown to contribute to air pollution disparities in multiple U.S. cities, $^{83-87}$ and their exhaust has been associated with a myriad of adverse health

effects. ¹⁸ Because HDDVs transport commercial goods, their emissions are reduced on weekends. ^{35,88} At the same time, passenger vehicle traffic and point source NO_x emissions do not exhibit similarly significant weekday—weekend decreases, ^{83,89} although the timing of these emissions shift with greater weekend traffic outside of rush hour at midday. ⁸⁹

To evaluate the influence of HDDV emissions on NO2 column observations, we compared annual weekday (Tuesday-Friday) and weekend (Saturday-Sunday) tract-level population-weighted TROPOMI NO₂ measurements (Figure 2b,c). NO₂ columns unweighted by population were 29% lower on weekends than on weekdays with weekend NO2 column enhancements primarily near the HSC and western shore of Galveston Bay (Figure 3c) where industrial sources in the HMA are concentrated. We calculated greater weekend NO_2 decreases for LINs (24 \pm 2%) than for HIWs (15 \pm 2%). If HDDV traffic caused the entire weekday-weekend difference, then HDDV emissions contributed 25 \pm 3% of the LIN-HIW population-weighted NO₂ column inequality across Houston. The same calculation based on inventory data produced weekday-weekend differences of $22 \pm 2\%$ with the slight variance compared to the observations, potentially due to co-varying gasoline vehicle traffic patterns and/or changes in the NO₂ chemical lifetime. Comparable results between TROPOMI and FIVE + NEI, at least in the HMA, are in part due to the uniquely high spatial resolution of the FIVE inventory (500 m × 500 m); for example, standard NEI emission files (that include mobile sources) are 4 km \times 4 km, typically too coarse to resolve traffic emissions along highways. 90 TROPOMI may therefore be suited to inform NO₂ inequality analyses in cities with incomplete, uncertain, out-of-date, and/or poorly-resolved (in space and time) inventories.

Conclusions and Looking Forward. We quantified the unequal distribution of NO₂ across the city of Houston using high spatial resolution GCAS airborne observations (250 m × 500 m), which were fine enough to resolve census-tract-scale NO₂ spatial variability. During DISCOVER-AQ, we found that population-weighted NO₂ was 37 ± 6% higher for non-whites and Hispanics living in low-income tracts compared to whites living in high-income tracts; additionally, there were NO2 disparities of 11-32% by race-ethnicity and 15-28% by poverty status. We used this analysis as a basis of comparison to determine that TROPOMI resolved similar tract-level NO2 disparities. We observed that, while population-weighted GCAS NO₂ columns were always greater for non-white, Hispanic, and low-income residents, they were also substantially more temporally variable, a consequence of the higher NO_x source density in these neighborhoods. Greater temporal variability has implications for research on the acute health impacts of NO₂, and its surrogates, for LINs, as time-averaged pollutant data and static LUR models do not represent high NO2 events, such as that observed on September 25 and reflected in the shift in the NO2 column statistical distribution from log-normal (GCAS, 35 flight circuits) to normal (TROPOMI, one year of weekends) (Figure 4). Because long-term averaging does not inherently change the underlying data distribution and given that TROPOMI has at least the potential for daily coverage, the impact of transient extreme events could still be captured by analyzing other aspects of the NO₂ column distribution. In addition, the multiyear time record of the TROPOMI and ongoing data collection provide empirical evidence of specific time-varying NO_x source

contributions potentially relevant for political and regulatory decision making; for example, weekday—weekend NO_2 differences in Houston indicate that HDDVs cause up to 25% of the city's NO_2 disparities. Such information can be used by city planners, elected officials, and other decision makers as they develop comprehensive plans, allocate resources for mitigation, invest in public transportation, and/or implement vehicle bans.

Spatially and temporally extensive air quality observations are foundational for successful policy design, implementation, and evaluation in every city, especially for controversial issues like environmental justice; this is because they can reduce uncertainties about the severity of disparities for specific chemicals, which are often at the root of related policy disputes. Here, we demonstrated that TROPOMI (oversampled to 0.01° × 0.01°) precisely observes NO₂ differences between census tracts and the spatial patterns in NO2 columns reflect those measured at the surface. However, our results also suggested that TROPOMI-derived NO2 disparities generally represent lower bounds with time averaging needed to improve measurement spatial resolution and documented retrieval biases that minimize absolute NO2 differences. Still, TROPOMI measurements may be well positioned to inform multiple aspects of city-wide decision making in novel ways, for example, in the development of local interventions, such as ordinances, moratoriums, comprehensive plans, zoning, siting decisions, traffic planning, and permitting; through data-driven regulatory enforcement; and by supporting resource prioritization for environmental equity, especially in areas lacking surface monitoring. Because NO_x partitioning favors NO in the nearfield of sources, which are more prevalent in non-white, Hispanic, and low-income neighborhoods, at least in Houston, broader application of satellite NO2 columns as a surrogate for toxic combustion emissions requires further consideration. Even so, the TROPOMI observations have the potential to promote targeted and tailored municipal NO_x intervention efforts, illuminate limitations to cities' enforcement capacities (e.g., growth in road transport), and inform policies to ensure compliance and equitable outcomes. Finally, the geostationary satellite TEMPO instrument (planned launch date in 2022) will provide hourly column observations of up to 2 km \times 4.5 km at the center of its field of regard over North America, resulting in the most spatially and temporally precise spacebased NO₂ observations over the United States to date. ⁹¹ Finer city-wide spatiotemporal details will further expand our ability to reveal, document, and monitor census-tract-level NO2 disparities to observe high-NO2 events, infer source contributions from temporal variability, and inform decision making to eliminate the practices, behaviors, and conditions contributing to racial, ethnic, and income-based air quality disparities in major U.S. cities.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.0c01864.

Study area maps, individual GCAS circuit NO₂ maps, surface wind statistics on GCAS flight days, temporal trends in surface NO₂*, TROPOMI columns subsampled along the GCAS track, NASA P-3B flight path and profile, CBL and in situ NO₂ column statistics, and

all individual measured NO_2 , theta lapse rate, theta, and $H_2O_{(v)}$ profiles (Figures S15–S63) (PDF)

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Notes

The authors declare no competing financial interest. Data files of GCAS and TROPOMI observations averaged to census tracts are available for download at https://pusede.evsc.virginia.edu/home/data/.

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REFERENCES

- (1) State of the Air 2019; American Lung Association: April 24, 2019.
- (2) National Air Toxics Assessment 2014; U.S. Environmental Protection Agency, 2014.
- (3) Collins, T. W.; Grineski, S. E.; Chakraborty, J.; Montgomery, M. C.; Hernandez, M. Downscaling Environmental Justice Analysis: Determinants of Household-Level Hazardous Air Pollutant Exposure in Greater Houston. *Ann. Am. Assoc. Geogr.* **2015**, *105*, 684–703.
- (4) Hernandez, M.; Collins, T. W.; Grineski, S. E. Immigration, mobility, and environmental injustice: A comparative study of Hispanic people's residential decision-making and exposure to hazardous air pollutants in Greater Houston, Texas. *Geoforum* **2015**, *60*, 83–94.
- (5) Bullard, R. D. Invisible Houston: the Black experience in boom and bust; Texas A & M University Press: College Station, 1987.
- (6) Linder, S. H.; Marko, D.; Sexton, K. Cumulative Cancer Risk from Air Pollution in Houston: Disparities in Risk Burden and Social Disadvantage. *Environ. Sci. Technol.* **2008**, 42, 4312–4322.
- (7) Sansom, G.; Parras, J.; Parras, A.; Nieto, Y.; Arellano, Y.; Berke, P.; McDonald, T.; Shipp, E.; Horney, J. A. The Impacts of Exposure to Environmental Risk on Physical and Mental Health in a Small Geographic Community in Houston, TX. *J. Community Health* **2017**, 42, 813–818.
- (8) Clark, L. P.; Millet, D. B.; Marshall, J. D. Changes in Transportation-Related Air Pollution Exposures by Race-Ethnicity and Socioeconomic Status: Outdoor Nitrogen Dioxide in the United States in 2000 and 2010. *Environ. Health Perspect.* **2017**, *125*, 097012.
- (9) Clark, L. P.; Millet, D. B.; Marshall, J. D. National Patterns in Environmental Injustice and Inequality: Outdoor NO₂ Air Pollution in the United States. *PLoS One* **2014**, *9*, No. e94431.
- (10) Levy, I.; Mihele, C.; Lu, G.; Narayan, J.; Brook, J. R. Evaluating Multipollutant Exposure and Urban Air Quality: Pollutant Interrelationships, Neighborhood Variability, and Nitrogen Dioxide as a Proxy Pollutant. *Environ. Health Perspect.* **2014**, *122*, 65–72.
- (11) Jerrett, M.; Burnett, R. T.; Ma, R.; Pope, C. A., III; Krewski, D.; Newbold, K. B.; Thurston, G.; Shi, Y.; Finkelstein, N.; Calle, E. E.; Thun, M. J. Spatial analysis of air pollution and mortality in Los Angeles. *Epidemiology* **2005**, *16*, 727.
- (12) Morello-Frosch, R.; Jesdale, B. M. Separate and unequal: residential segregation and estimated cancer risks associated with ambient air toxics in U.S. metropolitan areas. *Environ. Health Perspect.* **2006**, *114*, 386–393.
- (13) Hajat, A.; Hsia, C.; O'Neill, M. S. Socioeconomic Disparities and Air Pollution Exposure: a Global Review. *Curr. Environ. Health Rep.* **2015**, *2*, 440–450.
- (14) Profile of version 1 of the 2014 National Emissions Inventory; Office of Air Quality Planning and Standards, Emissions Inventory and Analysis Group: April, 2017.
- (15) Kim, S.-W.; McKeen, S. A.; Frost, G. J.; Lee, S.-H.; Trainer, M.; Richter, A.; Angevine, W. M.; Atlas, E.; Bianco, L.; Boersma, K. F.; Brioude, J.; Burrows, J. P.; de Gouw, J.; Fried, A.; Gleason, J. F.; Hilboll, A.; Mellqvist, J.; Peischl, J.; Richter, D.; Rivera, C.; Ryerson, T.; te Lintel Hekkert, S.; Walega, J.; Warneke, C.; Weibring, P.; Williams, E. Evaluations of NO_x and highly reactive VOC emission inventories in Texas and their implications for ozone plume simulations during the Texas Air Quality Study 2006. *Atmos. Chem. Phys.* **2011**, *11*, 11361–11386.
- (16) Rivera, C.; Mellqvist, J.; Samuelsson, J.; Lefer, B.; Alvarez, S.; Patel, M. R. Quantification of NO₂ and SO₂ emissions from the Houston Ship Channel and Texas City industrial areas during the 2006 Texas Air Quality Study. *J. Geophys. Res.: Atmos.* **2010**, 27, D08301.

- (17) Washenfelder, R. A.; Trainer, M.; Frost, G. J.; Ryerson, T. B.; Atlas, E. L.; de Gouw, J. A.; Flocke, F. M.; Fried, A.; Holloway, J. S.; Parrish, D. D.; Peischl, J.; Richter, D.; Schauffler, S. M.; Walega, J. G.; Warneke, C.; Weibring, P.; Zheng, W. Characterization of NO_x, SO₂, ethene, and propene from industrial emission sources in Houston, Texas. *J. Geophys. Res.: Atmos.* **2010**, *115*, D16311.
- (18) Traffic-Related Air Pollution: A Critical Review of the Literature on Emissions, Exposure, and Health Effects; Health Effects Institute, 2010, (Special Report 17).
- (19) Brook, J. R.; Burnett, R. T.; Dann, T. F.; Cakmak, S.; Goldberg, M. S.; Fan, X.; Wheeler, A. J. Further interpretation of the acute effect of nitrogen dioxide observed in Canadian time-series studies. *J. Exposure Sci. Environ. Epidemiol.* **2007**, *17*, S36—S44.
- (20) Brunekreef, B.; Holgate, S. T. Air pollution and health. *Lancet* **2002**, *360*, 1233–1242.
- (21) Burnett, R. T.; Stieb, D.; Brook, J. R.; Cakmak, S.; Dales, R.; Raizenne, M.; Vincent, R.; Dann, T. Associations between short-term changes in nitrogen dioxide and mortality in Canadian cities. *Arch. Environ. Health* **2004**, *59*, 228–236.
- (22) Crouse, D. L.; Peters, P. A.; Hystad, P.; Brook, J. R.; van Donkelaar, A.; Martin, R. V.; Villeneuve, P. J.; Jerrett, M.; Goldberg, M. S.; Pope, C. A., III; Brauer, M.; Brook, R. D.; Robichaud, A.; Menard, R.; Burnett, R. T. Ambient PM_{2.5}, O₃, and NO₂ Exposures and Associations with Mortality over 16 Years of Follow-Up in the Canadian Census Health and Environment Cohort (CanCHEC). *Environ. Health Perspect.* **2015**, *123*, 1180–1186.
- (23) Edwards, J.; Walters, S.; Griffiths, R. K. Hospital Admissions for Asthma in Preschool-Children Relationship to Major Roads in Birmingham, United-Kingdom. *Arch. Environ. Health* **1994**, 49, 223–227.
- (24) Gauderman, W. J.; Avol, E.; Lurmann, F.; Kuenzli, N.; Gilliland, F.; Peters, J.; McConnell, R. Childhood asthma and exposure to traffic and nitrogen dioxide. *Epidemiology* **2005**, *16*, 737–743.
- (25) Lin, S.; Munsie, J. P.; Hwang, S. A.; Fitzgerald, E.; Cayo, M. R. Childhood asthma hospitalization and residential exposure to state route traffic. *Environ. Res.* **2002**, *88*, 73–81.
- (26) Adar, S. D.; Kaufman, J. D. Cardiovascular disease and air pollutants: Evaluating and improving epidemiological data implicating traffic exposure. *Inhalation Toxicol.* **2007**, *19*, 135–149.
- (27) Lipfert, F. W.; Wyzga, R. E. On exposure and response relationships for health effects associated with exposure to vehicular traffic. *I. Exposure Sci. Environ. Epidemiol.* **2008**, *18*, 588–599.
- (28) Wu, J.; Ren, C.; Delfino, R. J.; Chung, J.; Wilhelm, M.; Ritz, B. Association between Local Traffic-Generated Air Pollution and Preeclampsia and Preterm Delivery in the South Coast Air Basin of California. *Environ. Health Perspect.* **2009**, *117*, 1773–1779.
- (29) Choi, W.; He, M.; Barbesant, V.; Kozawa, K. H.; Mara, S.; Winer, A. M.; Paulson, S. E. Prevalence of wide area impacts downwind of freeways under pre-sunrise stable atmospheric conditions. *Atmos. Environ.* **2012**, *62*, 318–327.
- (30) Karner, A. A.; Eisinger, D. S.; Niemeier, D. A. Near-Roadway Air Quality: Synthesizing the Findings from Real-World Data. *Environ. Sci. Technol.* **2010**, *44*, 5334–5344.
- (31) Bechle, M. J.; Millet, D. B.; Marshall, J. D. Remote sensing of exposure to NO₂: Satellite versus ground-based measurement in a large urban area. *Atmos. Environ.* **2013**, *69*, 345–353.
- (32) Rowangould, G. M. A census of the US near-roadway population: Public health and environmental justice considerations. *Trans. Res. D Trans. Environ.* **2013**, *25*, 59–67.
- (33) Boersma, K. F.; Jacob, D. J.; Trainic, M.; Rudich, Y.; DeSmedt, I.; Dirksen, R.; Eskes, H. J. Validation of urban NO₂ concentrations and their diurnal and seasonal variations observed from the SCIAMACHY and OMI sensors using in situ surface measurements in Israeli cities. *Atmos. Chem. Phys.* **2009**, *9*, 3867–3879.
- (34) Geddes, J. A.; Martin, R. V.; Boys, B. L.; van Donkelaar, A. Long-Term Trends Worldwide in Ambient NO₂ Concentrations Inferred from Satellite Observations. *Environ. Health Perspect.* **2016**, 124, 281–289.

- (35) Russell, A. R.; Valin, L. C.; Cohen, R. C. Trends in OMI NO₂ Observations over the United States: Effects of Emission Control Technology and the Economic Recession. *Atmos. Chem. Phys.* **2012**, 12, 12197–12209.
- (36) Judd, L. M.; Al-Saadi, J. A.; Janz, S. J.; Kowalewski, M. G.; Pierce, R. B.; Szykman, J. J.; Valin, L. C.; Swap, R.; Cede, A.; Mueller, M.; Tiefengraber, M.; Abuhassan, N.; Williams, D. Evaluating the impact of spatial resolution on tropospheric NO₂ column comparisons within urban areas using high-resolution airborne data. *Atmos. Meas. Tech.* **2019**, *12*, 6091–6111.
- (37) Valin, L. C.; Russell, A. R.; Bucsela, E. J.; Veefkind, J. P.; Cohen, R. C. Observation of slant column NO₂ using the super-zoom mode of AURA-OMI. *Atmos. Meas. Tech.* **2011**, *4*, 1929–1935.
- (38) Larkin, A.; Geddes, J. A.; Martin, R. V.; Xiao, Q.; Liu, Y.; Marshall, J. D.; Brauer, M.; Hystad, P. Global Land Use Regression Model for Nitrogen Dioxide Air Pollution. *Environ. Sci. Technol.* **2017**, *51*, 6957–6964.
- (39) Novotny, E. V.; Bechle, M. J.; Millet, D. B.; Marshall, J. D. National Satellite-Based Land-Use Regression: NO_2 in the United States. *Environ. Sci. Technol.* **2011**, *45*, 4407–4414.
- (40) Su, J. G.; Jerrett, M.; Beckerman, B.; Wilhelm, M.; Ghosh, J. K.; Ritz, B. Predicting traffic-related air pollution in Los Angeles using a distance decay regression selection strategy. *Environ. Res.* **2009**, *109*, 657–670.
- (41) Nowlan, C. R.; Liu, X.; Janz, S. J.; Kowalewski, M. G.; Chance, K.; Follette-Cook, M. B.; Fried, A.; González Abad, G.; Herman, J. R.; Judd, L. M.; Kwon, H.-A.; Loughner, C. P.; Pickering, K. E.; Richter, D.; Spinei, E.; Walega, J.; Weibring, P.; Weinheimer, A. J. Nitrogen dioxide and formaldehyde measurements from the GEOstationary Coastal and Air Pollution Events (GEO-CAPE) Airborne Simulator over Houston, Texas. *Atmos. Meas. Tech.* **2018**, *11*, 5941–5964.
- (42) Nowlan, C. R.; Liu, X.; Leitch, J. W.; Chance, K.; González Abad, G.; Liu, C.; Zoogman, P.; Cole, J.; Delker, T.; Good, W.; Murcray, F.; Ruppert, L.; Soo, D.; Follette-Cook, M. B.; Janz, S. J.; Kowalewski, M. G.; Loughner, C. P.; Pickering, K. E.; Herman, J. R.; Beaver, M. R.; Long, R. W.; Szykman, J. J.; Judd, L. M.; Kelley, P.; Luke, W. T.; Ren, X.; Al-Saadi, J. A. Nitrogen dioxide observations from the Geostationary Trace gas and Aerosol Sensor Optimization (GeoTASO) airborne instrument: Retrieval algorithm and measurements during DISCOVER-AQ Texas 2013. *Atmos. Meas. Tech.* 2016, 9, 2647–2668.
- (43) Souri, A. H.; Choi, Y.; Pan, S.; Curci, G.; Nowlan, C. R.; Janz, S. J.; Kowalewski, M. G.; Liu, J.; Herman, J. R.; Weinheimer, A. J. First Top-Down Estimates of Anthropogenic NOx Emissions Using High-Resolution Airborne Remote Sensing Observations. *J. Geophys. Res.: Atmos.* 2018, 123, 3269–3284.
- (44) U.S. Census Burea, P. D. Annual Estimates of Resident Population. April 1, 2010 to July 1, 2018. https://factfinder.census.gov/faces/tableservices/jsf/pages/productview.xhtml?src=bkmk (accessed August 1).
- (45) Texas State Energy Profile. https://www.eia.gov/state/print.php?sid=TX(accessed October 7, 2019).
- (46) Senate Select Committee on Texas Ports. Overview of Texas Ports and Waterways; May 3, 2016, 2016.
- (47) Darby, L. S. Cluster Analysis of Surface Winds in Houston, Texas, and the Impact of Wind Patterns on Ozone. *J. Appl. Meteorol.* **2005**, *44*, 1788–1806.
- (48) State of Global Air 2019; Health Effects Institute, Boston, MA, 2019.
- (49) Sexton, K.; Linder, S. H.; Marko, D.; Bethel, H.; Lupo, P. J. Comparative assessment of air pollution-related health risks in Houston. *Environ. Health Perspect.* **2007**, *115*, 1388–1393.
- (50) Leitch, J. W. D. T.; Good, W.; Ruppert, L.; Murcray, F.; Chance, K.; Liu, X.; Nowlan, C.; Janz, S. J.; Krotkov, N. A.; Pickering, K. E.; Kowalewski, M.; Wang, J. The GeoTASO airborne spectrometer project. In *Earth Observing Systems*; International Society for Optics and Photonics, p 9., 2014.
- (51) Skamarock, W. C.; Klemp, J. B.; Dudhia, J.; Gill, D. O.; Barker, D. M.; Duda, M. G.; Huang, X.-Y.; Wang, W.; Powers, J. G. A

- Description of the Advanced Research WRF Version 3; University Corporation for Atmospheric Research: Boulder, Colorado, 2008.
- (52) Otte, T. L.; Pleim, J. E. The Meteorology-Chemistry Interface Processor (MCIP) for the CMAQ modeling system: updates through MCIPv3.4.1. *Geosci. Model Dev.* **2010**, *3*, 243–256.
- (53) Loughner, C. P.; Follette-Cook, M. Emission source region contributions to a high surface ozone episode during DISCOVER-AQ, final Report to Texas AQRP; 2015.
- (54) van Geffen, J. H. G.; Boersma, K. F.; Eskes, H. J.; Maasakkers, J. D.; Veefkind, J. P. *TROPOMI ATBD of the total and tropospheric NO2 data products.* http://www.tropomi.eu (accessed 12 May).
- (55) Veefkind, J. P.; Aben, I.; McMullan, K.; Förster, H.; de Vries, J.; Otter, G.; Claas, J.; Eskes, H. J.; de Haan, J. F.; Kleipool, Q.; van Weele, M.; Hasekamp, O.; Hoogeveen, R.; Landgraf, J.; Snel, R.; Tol, P.; Ingmann, P.; Voors, R.; Kruizinga, B.; Vink, R.; Visser, H.; Levelt, P. F. TROPOMI on the ESA Sentinel-5 Precursor: A GMES mission for global observations of the atmospheric composition for climate, air quality and ozone layer applications. *Remote Sens. Environ.* **2012**, *120*, 70–83.
- (56) Boersma, K. F.; Eskes, H. J.; Dirksen, R. J.; van der A, R. J.; Veefkind, J. P.; Stammes, P.; Huijnen, V.; Kleipool, Q. L.; Sneep, M.; Claas, J.; Leitao, J.; Richter, A.; Zhou, Y.; Brunner, D. An improved tropospheric NO₂ column retrieval algorithm for the Ozone Monitoring Instrument. *Atmos. Meas.Tech.* **2011**, *4*, 1905–1928.
- (57) Lorente, A.; Folkert Boersma, K.; Yu, H.; Dörner, S.; Hilboll, A.; Richter, A.; Liu, M.; Lamsal, L. N.; Barkley, M.; De Smedt, I.; Van Roozendael, M.; Wang, Y.; Wagner, T.; Beirle, S.; Lin, J. T.; Krotkov, N.; Stammes, P.; Wang, P.; Eskes, H. J.; Krol, M. Structural uncertainty in air mass factor calculation for NO₂ and HCHO satellite retrievals. *Atmos. Meas. Tech.* **2017**, *10*, 759–782.
- (58) van Geffen, J. H. G. M.; Boersma, K. F.; Van Roozendael, M.; Hendrick, F.; Mahieu, E.; De Smedt, I.; Sneep, M.; Veefkind, J. P. Improved spectral fitting of nitrogen dioxide from OMI in the 405–465 nm window. *Atmos. Meas. Tech.* **2015**, *8*, 1685–1699.
- (59) Zara, M.; Boersma, K. F.; De Smedt, I.; Richter, A.; Peters, E.; van Geffen, J. H. G. M.; Beirle, S.; Wagner, T.; Van Roozendael, M.; Marchenko, S.; Lamsal, L. N.; Eskes, H. J. Improved slant column density retrieval of nitrogen dioxide and formaldehyde for OMI and GOME-2A from QA4ECV: intercomparison, uncertainty characterisation, and trends. *Atmos. Meas. Tech.* 2018, 11, 4033–4058.
- (60) Boersma, K. F.; Eskes, H. J.; Richter, A.; De Smedt, I.; Lorente, A.; Beirle, S.; van Geffen, J. H. G. M.; Zara, M.; Peters, E.; Van Roozendael, M.; Wagner, T.; Maasakkers, J. D.; van der A, R. J.; Nightingale, J.; De Rudder, A.; Irie, H.; Pinardi, G.; Lambert, J. C.; Compernolle, S. C. Improving algorithms and uncertainty estimates for satellite NO₂ retrievals: results from the quality assurance for the essential climate variables (QA4ECV) project. *Atmos. Meas.Tech.* **2018**, *11*, 6651–6678.
- (61) Boersma, K. F.; Eskes, H. J.; Brinksma, E. J. Error analysis for tropospheric NO₂ retrieval from space. *J. Geophys. Res.: Atmos.* **2004**, 109, D04311.
- (62) Williams, J. E.; Boersma, K. F.; Le Sager, P.; Verstraeten, W. W. The high-resolution version of TM5-MP for optimized satellite retrievals: description and validation. *Geosci. Model Dev.* **2017**, *10*, 721–750.
- (63) Kleipool, Q. L.; Dobber, M. R.; de Haan, J. F.; Levelt, P. F. Earth surface reflectance climatology from 3 years of OMI data. *J. Geophys. Res.-Atmos.* **2008**, *113*, D18308.
- (64) Eskes, H.; Geffen, J. V.; Boersma, F.; Eichmann, K.-U.; Apituley, A.; Pedergnana, M.; Sneep, M.; Veefkind, J. P.; Loyola, D. Sentinel-5 precursor/TROPOMI Level 2 Product User Manual Nitrogendioxide; Ministry of Infrastructure and Water Management: 2019.
- (65) Sun, K.; Zhu, L.; Cady-Pereira, K.; Chan Miller, C.; Chance, K.; Clarisse, L.; Coheur, P. F.; González Abad, G.; Huang, G.; Liu, X.; Van Damme, M.; Yang, K.; Zondlo, M. A physics-based approach to oversample multi-satellite, multispecies observations to a common grid. *Atmos. Meas. Tech.* **2018**, *11*, 6679–6701.
- (66) Dunlea, E. J.; Herndon, S. C.; Nelson, D. D.; Volkamer, R. M.; San Martini, F.; Sheehy, P. M.; Zahniser, M. S.; Shorter, J. H.;

- Wormhoudt, J. C.; Lamb, B. K.; Allwine, E. J.; Gaffney, J. S.; Marley, N. A.; Grutter, M.; Marquez, C.; Blanco, S.; Cardenas, B.; Retama, A.; Ramos Villegas, C. R.; Kolb, C. E.; Molina, L. T.; Molina, M. J. Evaluation of nitrogen dioxide chemiluminescence monitors in a polluted urban environment. *Atmos. Chem. Phys.* **2007**, *7*, 2691–2704. (67) Russell, A. R.; Valin, L. C.; Bucsela, E. J.; Wenig, M. O.; Cohen,
- (67) Russell, A. R.; Valin, L. C.; Bucsela, E. J.; Wenig, M. O.; Cohen, R. C. Space-based Constraints on Spatial and Temporal Patterns of NO_x Emissions in California, 2005–2008. *Environ. Sci. Technol.* **2010**, 44, 3608–3615.
- (68) Ambient Monitoring Technology Information Center (AMTIC), Environmental Protection agency, Near-road Monitoring Sites (Excel), Near-road NO2 Monitoring. (accessed September 15).
- (69) Manson, S.; Shroeder, J.; Van Riper, D.; Ruggles, S. IPUMS National Historical Geographic Information System: Version 13.0; Minneapolis: University of Minnesota, 2018.
- (70) Hennemuth, B.; Lammert, A. Determination of the Atmospheric Boundary Layer Height from Radiosonde and Lidar Backscatter. *Boundary Layer Meteorol.* **2006**, *120*, 181–200.
- (71) Dai, C.; Wang, Q.; Kalogiros, J. A.; Lenschow, D. H.; Gao, Z.; Zhou, M. Determining Boundary-Layer Height from Aircraft Measurements. *Boundary Layer Meteorol.* **2014**, *152*, 277–302.
- (72) McDonald, B. C.; McKeen, S. A.; Cui, Y. Y.; Ahmadov, R.; Kim, S.-W.; Frost, G. J.; Pollack, I. B.; Peischl, J.; Ryerson, T. B.; Holloway, J. S.; Graus, M.; Warneke, C.; Gilman, J. B.; de Gouw, J. A.; Kaiser, J.; Keutsch, F. N.; Hanisco, T. F.; Wolfe, G. M.; Trainer, M. Modeling Ozone in the Eastern U.S. using a Fuel-Based Mobile Source Emissions Inventory. *Environ. Sci. Technol.* **2018**, *52*, 7360–7370.
- (73) McDonald, B. C.; Dallmann, T. R.; Martin, E. W.; Harley, R. A. Long-term trends in nitrogen oxide emissions from motor vehicles at national, state, and air basin scales. *J. Geophys. Res.-Atmos.* **2012**, *117*, D00V18
- (74) Frost, G. J.; McKeen, S. A.; Trainer, M.; Ryerson, T. B.; Neuman, J. A.; Roberts, J. M.; Swanson, A.; Holloway, J. S.; Sueper, D. T.; Fortin, T.; Parrish, D. D.; Fehsenfeld, F. C.; Flocke, F.; Peckham, S. E.; Grell, G. A.; Kowal, D.; Cartwright, J.; Auerbach, N.; Habermann, T. Effects of changing power plant NO_x emissions on ozone in the eastern United States: Proof of concept. *J. Geophys. Res.-Atmos.* **2006**, *111*, D12306.
- (75) Jiang, Z.; McDonald, B. C.; Worden, H.; Worden, J. R.; Miyazaki, K.; Qu, Z.; Henze, D. K.; Jones, D. B. A.; Arellano, A. F.; Fischer, E. V.; Zhu, L.; Boersma, K. F. Unexpected slowdown of US pollutant emission reduction in the past decade. *Proc. Natl. Acad. Sci. U. S. A.* 2018, 115, 5099–5104.
- (76) Judd, L. M.; Al-Saadi, J. A.; Szykman, J. J.; Valin, L. C.; Janz, S. J.; Kowalewski, M. G.; Eskes, H. J.; Veefkind, J. P.; Cede, A.; Mueller, M.; Gebetsberger, M.; Swap, R.; Pierce, R. B.; Nowlan, C. R.; Abad, G. G.; Nehrir, A.; Williams, D. Evaluating Sentinel-5P TROPOMI tropospheric NO₂ column densities with airborne and Pandora spectrometers near New York City and Long Island Sound. *Atmos. Meas. Tech. Discuss.* **2020**, 2020, 1–52.
- (77) Griffin, D.; Zhao, X.; McLinden, C. A.; Boersma, F.; Bourassa, A.; Dammers, E.; Degenstein, D.; Eskes, H.; Fehr, L.; Fioletov, V.; Hayden, K.; Kharol, S. K.; Li, S.-M.; Makar, P.; Martin, R. V.; Mihele, C.; Mittermeier, R. L.; Krotkov, N.; Sneep, M.; Lamsal, L. N.; Linden, M. t.; van Geffen, J.; Veefkind, P.; Wolde, M. High-Resolution Mapping of Nitrogen Dioxide With TROPOMI: First Results and Validation Over the Canadian Oil Sands. *Geophys. Res. Lett.* **2019**, *46*, 1049–1060.
- (78) Pope, R. J.; Graham, A. M.; Chipperfield, M. P.; Veefkind, J. P. High resolution satellite observations give new view of UK air quality. *Weather* **2019**, *74*, 316–320.
- (79) Goldberg, D. L.; Lu, Z.; Streets, D. G.; de Foy, B.; Griffin, D.; McLinden, C. A.; Lamsal, L. N.; Krotkov, N. A.; Eskes, H. Enhanced Capabilities of TROPOMI NO₂: Estimating NO_x from North American Cities and Power Plants. *Environ. Sci. Technol.* **2019**, 12594.
- (80) Ialongo, I.; Virta, H.; Eskes, H.; Hovila, J.; Douros, J. Comparison of TROPOMI/Sentinel 5 Precursor NO₂ observations with ground-based measurements in Helsinki. *Atmos. Meas. Tech.* **2020**, *13*, 205–218.

- (81) McLinden, C. A.; Fioletov, V.; Boersma, K. F.; Kharol, S. K.; Krotkov, N.; Lamsal, L.; Makar, P. A.; Martin, R. V.; Veefkind, J. P.; Yang, K. Improved satellite retrievals of NO₂ and SO₂ over the Canadian oil sands and comparisons with surface measurements. *Atmos. Chem. Phys.* **2014**, *14*, 3637–3656.
- (82) Zhang, Y.; Wang, Y.; Chen, G.; Smeltzer, C.; Crawford, J.; Olson, J.; Szykman, J.; Weinheimer, A. J.; Knapp, D. J.; Montzka, D. D.; Wisthaler, A.; Mikoviny, T.; Fried, A.; Diskin, G. Large vertical gradient of reactive nitrogen oxides in the boundary layer: Modeling analysis of DISCOVER-AQ 2011 observations. *J. Geophys. Res.: Atmos.* 2016, 121, 1922–1934.
- (83) Houston, D.; Li, W.; Wu, J. Disparities in Exposure to Automobile and Truck Traffic and Vehicle Emissions Near the Los Angeles—Long Beach Port Complex. *Am. J. Public Health* **2014**, *104*, 156—164.
- (84) Houston, D.; Wu, J.; Ong, P.; Winer, A. Structural disparities of urban traffic in Southern California: Implications for vehicle-related air pollution exposure in minority and high-poverty neighborhoods. *J. Urban Affairs* **2004**, *26*, 565–592.
- (85) Houston, D.; Krudysz, M.; Winer, A. Diesel Truck Traffic in Low-Income and Minority Communities Adjacent to Ports:Environmental Justice Implications of Near-Roadway Land Use Conflicts. *Transp. Res. Rec.* **2008**, 2067, 38–46.
- (86) Lena, T. S.; Ochieng, V.; Carter, M.; Holguín-Veras, J.; Kinney, P. L. Elemental carbon and PM_{2.5} levels in an urban community heavily impacted by truck traffic. *Environ. Environ. Health Perspect.* **2002**, *110*, 1009–1015.
- (87) Karner, A.; Eisinger, D.; Bai, S.; Niemeier, D. Mitigating Diesel Truck Impacts in Environmental Justice Communities:Transportation Planning and Air Quality in Barrio Logan, San Diego, *California*. *Transp. Res. Rec.* **2009**, *2125*, 1–8.
- (88) Marr, L. C.; Harley, R. A. Modeling the Effect of Weekday-Weekend Differences in Motor Vehicle Emissions on Photochemical Air Pollution in Central California. *Environ. Sci. Technol.* **2002**, *36*, 4099–4106.
- (89) Nassar, R.; Napier-Linton, L.; Gurney, K. R.; Andres, R. J.; Oda, T.; Vogel, F. R.; Deng, F. Improving the temporal and spatial distribution of CO₂ emissions from global fossil fuel emission data sets. *J. Geophys. Res.: Atmos.* **2013**, *118*, 917–933.
- (90) McDonald, B. C.; McBride, Z. C.; Martin, E. W.; Harley, R. A. High-resolution mapping of motor vehicle carbon dioxide emissions. *J. Geophys. Res.: Atmos.* **2014**, *119*, 5283–5298.
- (91) Zoogman, P.; Liu, X.; Suleiman, R. M.; Pennington, W. F.; Flittner, D. E.; Al-Saadi, J. A.; Hilton, B. B.; Nicks, D. K.; Newchurch, M. J.; Carr, J. L.; Janz, S. J.; Andraschko, M. R.; Arola, A.; Baker, B. D.; Canova, B. P.; Chan Miller, C.; Cohen, R. C.; Davis, J. E.; Dussault, M. E.; Edwards, D. P.; Fishman, J.; Ghulam, A.; González Abad, G.; Grutter, M.; Herman, J. R.; Houck, J.; Jacob, D. J.; Joiner, J.; Kerridge, B. J.; Kim, J.; Krotkov, N. A.; Lamsal, L.; Li, C.; Lindfors, A.; Martin, R. V.; McElroy, C. T.; McLinden, C.; Natraj, V.; Neil, D. O.; Nowlan, C. R.; OSullivan, E. J.; Palmer, P. I.; Pierce, R. B.; Pippin, M. R.; Saiz-Lopez, A.; Spurr, R. J. D.; Szykman, J. J.; Torres, O.; Veefkind, J. P.; Veihelmann, B.; Wang, H.; Wang, J.; Chance, K. Tropospheric emissions: Monitoring of pollution (TEMPO). J. Quant. Spectroscopy Rad. Transfer 2017, 186, 17–39.