

1 Global NO₂ Changes Between 2019 and 2024 as Observed by 2 TROPOMI in Urban Areas and Emerging Hotspots

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10 **Abstract.** We present a global assessment of space-based urban nitrogen dioxide (NO₂) observations from 2019 to 2024 using
11 annual and monthly mean tropospheric vertical column densities (VCDs) from the TROPOspheric Monitoring Instrument
12 (TROPOMI). Across 11,500 cities defined by the Global Human Settlement Layer-Settlement Model (GHS-SMOD), we find
13 population-weighted annual mean urban NO₂ VCDs were lower in 2024 than 2019 in Europe (-13%) and Asia and Oceania (-
14 17%), with seasonal decomposition indicating that annual changes are largely driven by concentration decreases during
15 November-March. Aggregated urban VCD changes in North America, South America and Africa were statistically
16 insignificant, though numerous individual cities exhibited significant changes. Of larger cities, Tehran had the largest annual
17 mean NO₂ VCD ($>30 \times 10^{15}$ molecules cm⁻²) and Seoul experienced the largest reduction ($-9.4 \pm 1.0\%$ yr⁻¹; $p < 0.001$). We then
18 calculate NO₂ VCD urban enhancements (VCD_{ENH}) by removing background concentrations from urban signatures and
19 compare VCD_{ENH} to changes in nitrogen oxide (NO_x) emissions from two emissions inventories, highlighting regions with
20 potential inventory discrepancies. We find VCD_{ENH} changes exceed changes in inventory NO_x emissions in Europe, North
21 America and Asia and Oceania, with worse agreement in the Global South. We further identify changes in NO₂ near fossil fuel
22 operations and note conflict-related changes in NO₂, highlighting the responsiveness of satellite NO₂ to certain societal
23 disruptions. This work demonstrates the value in space-based remote sensing being an accountability agent for air pollution
24 emissions on a global scale and to identify changes in NO₂ in otherwise unmonitored regions.

25 1 Introduction

26 Nitrogen dioxide (NO₂) is a harmful air pollutant that originates from both anthropogenic and natural emissions sources,
27 including fossil fuel combustion, biomass burning, lightning, and soils (Dix et al., 2020; Jin et al., 2021; Schuman & Huntrieser,
28 2007; Huber et al., 2024), with fossil fuel combustion accounting for ~45% of total global nitrogen oxide emissions (Song et
29 al., 2021). Only a small amount of NO₂ is emitted from these sources directly, with nitric oxide (NO) being the primary
30 emissions product that quickly cycles to NO₂ in the presence of oxidants such as ozone (O₃) or peroxy radicals (HO₂ or RO₂).

31 The summed concentrations of NO and NO₂ are referred to as nitrogen oxides (NO_x = NO + NO₂), as the concentrations of
32 NO and NO₂ are inherently linked. NO₂ is more commonly targeted by regulatory measures than NO, as it constitutes the
33 majority of atmospheric NO_x concentrations and is linked to increased morbidity and mortality from long-term exposure,
34 particularly within urban environments (Chen et al., 2024). While NO_x is commonly associated with health risks, the direct
35 association between NO_x exposure and adverse health outcomes remains uncertain (Anenberg et al., 2022). Despite this, NO_x
36 contributes to known harmful secondary pollutants, including O₃ and fine particulate matter.

37 NO₂ concentrations are measured using: (1) in-situ monitoring, e.g. chemiluminescence analyzers at the surface, or (2) remote
38 sensing instrumentation leveraging the unique spectral properties of NO₂, that absorbs light most efficiently in the visible
39 portions (405 – 465 nm) of the electromagnetic spectrum (Lamsal et al., 2015). The latter method relies upon spectrometers
40 detecting in the UV-Visible spectral range to infer NO₂ vertical column densities (VCDs), defined as the summed concentration
41 of NO₂ in a column from the surface to an upper limit of the atmosphere, with the tropopause often used as the upper limit.
42 Spectrometers have been used to measure NO₂ VCDs from ground-level directed upward, from aircraft directed downward, or
43 from space-based satellites directed downward, including from the TROPOspheric Monitoring Instrument (TROPOMI)
44 onboard the Sentinel-5P satellite (Herman et al., 2009; Fishman et al., 2012; Veeffkind et al., 2012). NO₂ can also be remotely
45 sensed from ground-based instruments capable of inferring vertical profiles of NO₂, such as using multi-axis differential optical
46 absorption spectroscopy (MAX-DOAS; Vlemmix et al., 2010).

47 The earliest space-based spectrometers detecting NO₂ were flown on low-earth polar orbiting satellites and were launched
48 within the mid-1990s to mid-2000s. These include the Global Ozone Monitoring Experiment (GOME; Burrows et al., 1999)
49 and GOME-2 satellites, the SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY (SCIAMACHY;
50 Bovensmann et al., 1999) and the Ozone Monitoring Instrument (OMI; Levelt et al., 2006). The data collected using these
51 instruments provided unique insight into atmospheric chemistry and composition across the globe, including in mostly
52 unmonitored regions. OMI, launched in 2004, provided NO₂ VCDs at a spatial resolution of 13 × 24 km² at nadir and has
53 remained operable for more than two decades at the time this was written, providing a valuable long-term record of NO₂
54 globally. OMI remained the highest resolution space-based NO₂ product until TROPOMI launched in 2017, which ultimately
55 provided NO₂ VCDs at a spatial resolution of 3.5 × 5.5 km² at nadir. Observations at this resolution facilitated the evaluation
56 of satellite NO₂ at previously unprecedented spatial scales, including at the intra-urban level (Goldberg et al., 2021; Goldberg
57 et al., 2024).

58 NO₂ trends have been characterized in urban and broader environments using space-based instruments. Earlier satellite studies
59 used the GOME and SCIAMACHY satellites to identify increasing NO₂ VCD trends in China from the mid-1990s to the mid-
60 2000s (Richter et al., 2005; Stavrou et al., 2008; Van der A et al., 2008), driven primarily by economic growth and
61 industrialization. Later studies, incorporating OMI observations, highlighted further increases in China through the early
62 2010s, with VCDs and satellite-inferred surface concentrations steadily declining since (Miyazaki et al., 2017; Wang et al.,
63 2019; Jiang et al., 2022). Europe has exhibited steady NO₂ VCD declines since the start of the satellite NO₂ record (Richter et

64 al., 2005; Krotkov et al., 2016; Duncan et al., 2016), driven largely by the implementation of various emissions control
65 technologies. In the United States, NO₂ concentrations generally exhibited a decreasing trend from 2005 through the mid-
66 2010s (Lamsal et al., 2015), with VCD decreases more gradual since, in part due to an increased influence from regional
67 background NO₂ levels (Jiang et al., 2018; Goldberg et al., 2021; Dang et al., 2023). In contrast, urban regions of India have
68 shown NO₂ increases over the past few decades, linked to urbanization and energy demand growth (Hilboll et al., 2013; Ghude
69 et al., 2020). Over Africa and South America, NO₂ VCD trends through the mid-2010s have been less pronounced, reflecting
70 limited industrialization and more dominant contributions from biomass burning and natural sources (Geddes et al., 2016;
71 Castellanos et al., 2014). Additionally, numerous studies have highlighted the influence that the COVID-19 pandemic had on
72 NO₂ globally, with most regions globally exhibiting broad NO₂ decreases in 2020 during numerous lockdowns and subsequent,
73 regionally-distinct rebounds in emissions (Lonsdale et al., 2023; Fisher et al., 2024).

74 Satellite studies have been used to characterize trends within the urban environment specifically, using different methods to
75 characterize the urban extent. Geddes et al. (2016) used GOME, SCIAMACHY and GOME-2 oversampled to a $0.1^\circ \times 0.1^\circ$
76 grid to highlight NO₂ VCD trends globally, as well as in select urban areas, with the urban region defined as the surrounding
77 $\sim 200 \text{ km} \times 200 \text{ km}$. Fioletov et al. (2022) and Fioletov et al. (2025) used urban density from the Gridded Population of the
78 World (SEDAC, 2017) as a proxy for the extent of the urban environment to identify changes in urban NO_x emissions.
79 Anenberg et al. (2022) used urban boundaries provided from the 2019 version of the Global Human Settlement Layer-
80 Settlement model (GHS-SMOD) to evaluate NO₂ trends from 2000 – 2019 using surface NO₂ estimates derived from OMI
81 NO₂ and a land-use regression model.

82 Here, we use TROPOMI tropospheric NO₂ VCDs to quantify general NO₂ changes globally from 2019 to 2024, with a
83 particular focus on urban areas. The urban boundaries are defined by the 2023 version of GHS-SMOD, which provides urban
84 cluster boundaries for all urban regions globally. We evaluate changes in annual mean urban NO₂ VCDs against NO_x emissions
85 inventories and characterize the influence of different seasons on annual variations. We additionally note changes in select oil,
86 gas, and other mining regions, which exhibit the largest changes globally outside of urban areas. This study represents the first
87 detailed global-scale analysis of urban TROPOMI NO₂ from 2019 to 2024. Our findings illustrate how NO₂ responded to
88 specific societal events during this timeframe, such as the impact of clean air policies, population migration away from urban
89 areas due to war, the increased demand for fossil fuels and rare-Earth minerals, and the emergence and waning of a global
90 pandemic. Furthermore, by directly linking observed NO₂ urban enhancements with NO_x emission inventory data from the
91 updated EDGARv8.1, our work provides valuable insights into regions where emissions inventories align closely with
92 observations, as well as areas exhibiting potential inventory discrepancies. This work underscores the critical value of satellite-
93 derived NO₂ as a tool for urban air quality assessment and emissions management.

94 2 Data and Methods

95 2.1 Global Human Settlement Layer Urban Cluster Boundaries

96 The Global Human Settlement Layer-Settlement Model (GHS-SMOD; Schiavina et al., 2023) is a dataset developed by the
97 Joint Research Centre of the European Commission containing spatial boundaries and population estimates for all urban areas
98 globally with a population of at least 50,000, which can be used to subset gridded or spatially-disaggregated data for any built-
99 up area on Earth. GHS-SMOD uses satellite remote sensing to identify the spatial extent and boundaries of all cohesive built-
100 up areas globally at a spatial resolution of $1 \times 1 \text{ km}^2$, with each separate, cohesive built-up area referred to as an “urban cluster”.
101 In this study, we use the terms “urban cluster” and “city” interchangeably, although we note that GHS-SMOD urban clusters
102 do not always align with administrative city boundaries. GHS-SMOD has the benefit of providing a globally consistent,
103 satellite-derived definition of built-up areas, whereas administrative boundaries vary widely in definition and availability.
104 Using physical built-up area boundaries from GHS-SMOD instead of administrative ones may shift the absolute spatial extent
105 of some cities, but it does not materially alter the concentrations calculated in this study.

106 The 2023 version of GHS-SMOD provides boundaries for approximately 11,500 urban clusters, along with population
107 estimates for the year 2020 (Fig. S8). We note that GHS-SMOD urban clusters do not reflect the traditional boundaries of
108 individual cities as we may understand them, and as such, GHS-SMOD urban clusters can span multiple cities, regions or even
109 countries. For example, the urban cluster encompassing San Diego, California includes the city of San Diego, but also the
110 adjacent surrounding suburbs, as well as the entirety of Tijuana, Mexico (Fig. S9). In such cases, attribution of an urban cluster
111 to one particular city is not possible.

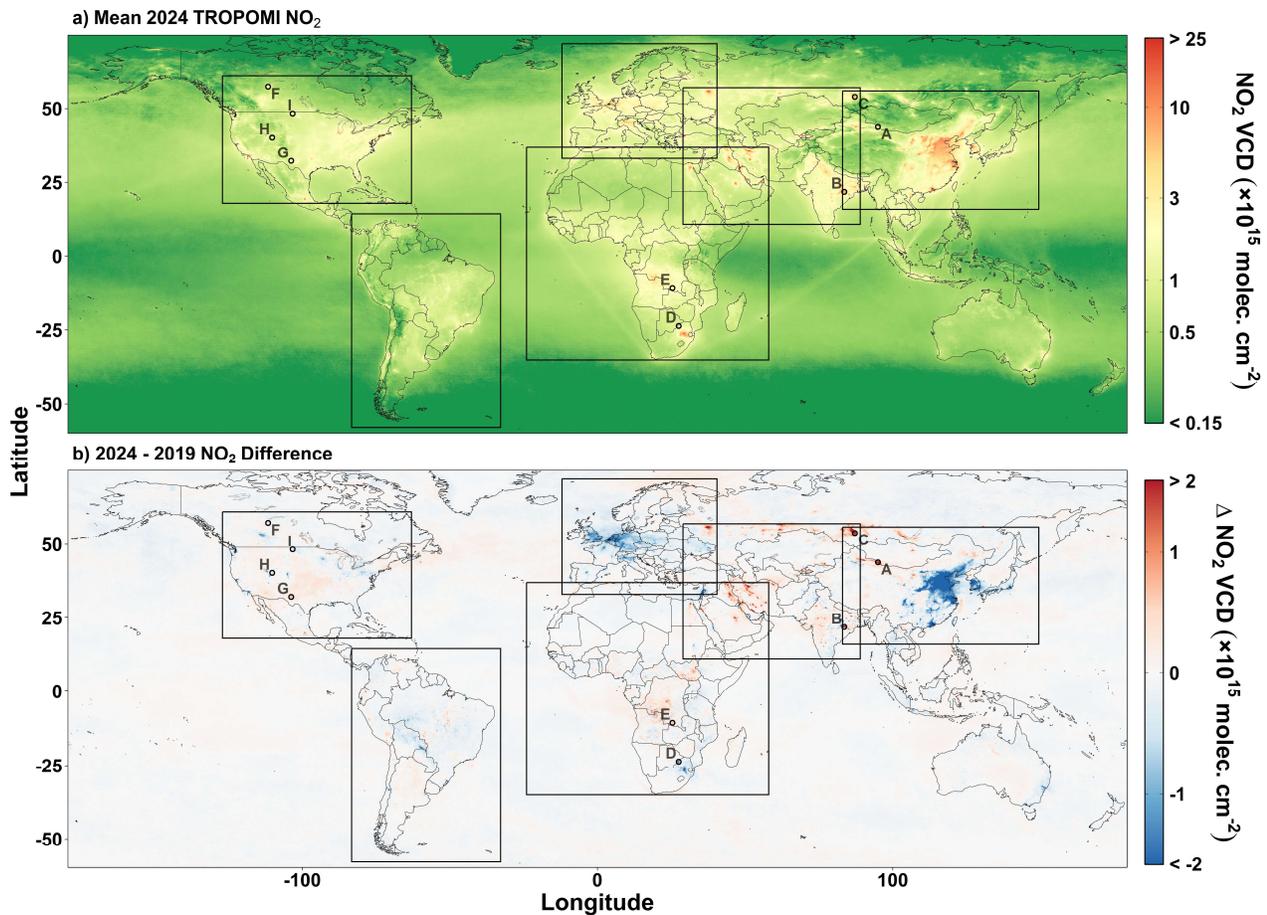
112 We use the GHS-SMOD boundaries to subset monthly- and annually-averaged satellite NO_2 column concentration data for all
113 urban clusters, as described in Section 2.2.1.

114 2.2 TROPOMI NO_2 Vertical Column Densities

115 The TROPOspheric Monitoring Instrument (TROPOMI) is a pushbroom spectrometer on board the Sentinel-5P satellite
116 traveling in low earth orbit, with approximately one overpass each afternoon (Veefkind et al., 2012). Launched in October
117 2017, TROPOMI detects radiation in spectral bands ranging from the ultraviolet to shortwave infrared to infer concentrations
118 of various atmospheric constituents, including nitrogen dioxide (NO_2), which is best inferred from the near-UV and visible
119 portions of the spectrum. We use Level 3 monthly- and annually-averaged TROPOMI tropospheric NO_2 vertical column
120 densities (VCDs) on a 0.1° global grid (Goldberg, 2024), which were created by oversampling daily Level 2 TROPOMI NO_2
121 VCDs derived from version 2.4+ of the European Space Agency retrieval algorithm (van Geffen et al., 2022). These Level 2
122 data have a nadir spatial resolution of $3.5 \times 7.0 \text{ km}^2$ before and $3.5 \times 5.5 \text{ km}^2$ after August 06, 2019. Data were quality
123 controlled to remove Level 2 pixels with a $qa_value < 0.75$ before oversampling, which removes data with quality issues
124 related to clouds, surface reflectivity (e.g. snow and ice) or other retrieval errors. The TROPOMI NO_2 data used in this study

125 span six full calendar years from January 2019 to December 2024 (Fig. 1); we use the RPRO version from 1 January 2019 –
126 25 July 2022 and the OFFL version from 26 July 2022 – 31 December 2024. On 7 September 2024 there was an update of the
127 surface reflectivity assumptions and on 16 November 2024 there was an update to the cloud retrieval, both of which induce a
128 small positive step change in the data but likely does not meaningfully affect the 2024 annual average.

129 TROPOMI NO₂ retrievals are subject to measurement and retrieval uncertainties that propagate into the oversampled Level 3
130 products. Typical uncertainties in monthly or annually averaged tropospheric NO₂ vertical column densities are on the order
131 of 15–20 %. Systematic biases have also been reported, with overestimation in less polluted regions (+26.5% bias) and
132 underestimation in areas with high NO₂ concentrations (-31.4% bias), reflecting limitations in the retrieval process (Glissenaar
133 et al., 2025; Lambert et al., 2025).



134
135 **Figure 1: (a) Global 2024 annual average NO₂ VCDs colored on a log-scale and (b) the difference in VCD from 2019 to 2024 colored**
136 **on a symmetric log-scale. Points labeled A-I correspond with locations of oil, gas and mining operations highlighted in Fig. 12. Boxes**
137 **indicate select focus regions in Section 5.**

138 2.3 Quantifying Average TROPOMI NO₂ VCDs for GHS-SMOD Urban Clusters

139 For each urban cluster, we subset the oversampled TROPOMI data for grid cells that are located within 0.1° of the urban
140 cluster boundary. For most cities, this results in approximately 20-25 grid cells, depending on the extent of the individual
141 cluster. Given that the spatial resolution of GHS-SMOD is roughly an order of magnitude finer than that of the oversampled
142 TROPOMI data (1 km vs. 0.1°) we interpolate the subsetted TROPOMI data to the 0.01° × 0.01° resolution of GHS-SMOD
143 using a nearest neighbor approach. We then calculate an area-weighted average of interpolated grid cells that have a grid cell
144 center falling within the urban cluster boundary (Fig. S9). This approach allows for the portions of oversampled 0.1° × 0.1°
145 grid cells that may not be centered within an urban cluster boundary, but that still overlap with a cluster, to be accounted for
146 within the average NO₂ column estimate.

147 To evaluate the changes in VCDs for broader regions, e.g. countries containing multiple urban clusters, we can calculate a
148 population-weighted average VCD, taking into account varying population sizes in different urban clusters.

$$149 \quad VCD_{PW} = \frac{\sum_i(POP_i \times VCD_i)}{\sum_i(POP_i)}, \quad (1)$$

150 In Eq. 1, VCD_{PW} represents the population-weighted VCD for a given country, POP_{*i*} represents the 2020 GHS-SMOD-
151 estimated population for a given urban cluster *i*, and VCD_{*i*} represents the mean NO₂ VCD for *i*.

152 For each time series, we use monthly TROPOMI NO₂ columns from 2019–2024 to estimate a change in % yr⁻¹. We first
153 construct a de-seasonalized anomaly series by computing, for each calendar month at each location, the mean NO₂ over the
154 full period and expressing each monthly value as a percent deviation from its corresponding monthly mean. To obtain the
155 percent change per year and its standard error, we fit a linear regression to the original monthly series with time as the predictor
156 and fixed effects for calendar month to control for seasonality. The estimated annual percent change and its standard error
157 were taken directly from the time-slope coefficient and its standard error from this regression. To assess statistical significance,
158 we regressed the de-seasonalized percent anomalies on time and obtained a p-value for the slope using standard errors that
159 account for temporal autocorrelation.

160 2.4 Accounting for Background NO₂

161 To account for changes in upwind background NO₂ concentrations that may influence urban NO₂ VCDs, we quantify an urban
162 NO₂ enhancement.

$$163 \quad VCD_{ENH} = VCD_{UC} - VCD_{BG}, \quad (2)$$

164 In Eq. 2, VCD_{ENH} is the urban NO₂ VCD enhancement, VCD_{UC} is the NO₂ VCD within each urban cluster as described in
165 Section 2.2.1, and VCD_{BG} is the background concentration for an urban cluster. We define VCD_{BG} for a given year as the 50th
166 percentile of annual mean NO₂ VCDs extending 0.5 degrees in any direction from an urban cluster boundary. Previous studies

167 have used a percentile threshold to determine background concentrations (de Gouw et al., 2020). See Section S1 of the
168 supplementary document for additional information and sensitivity tests regarding background VCD quantification.

169 **2.5 NO_x Emission Inventories**

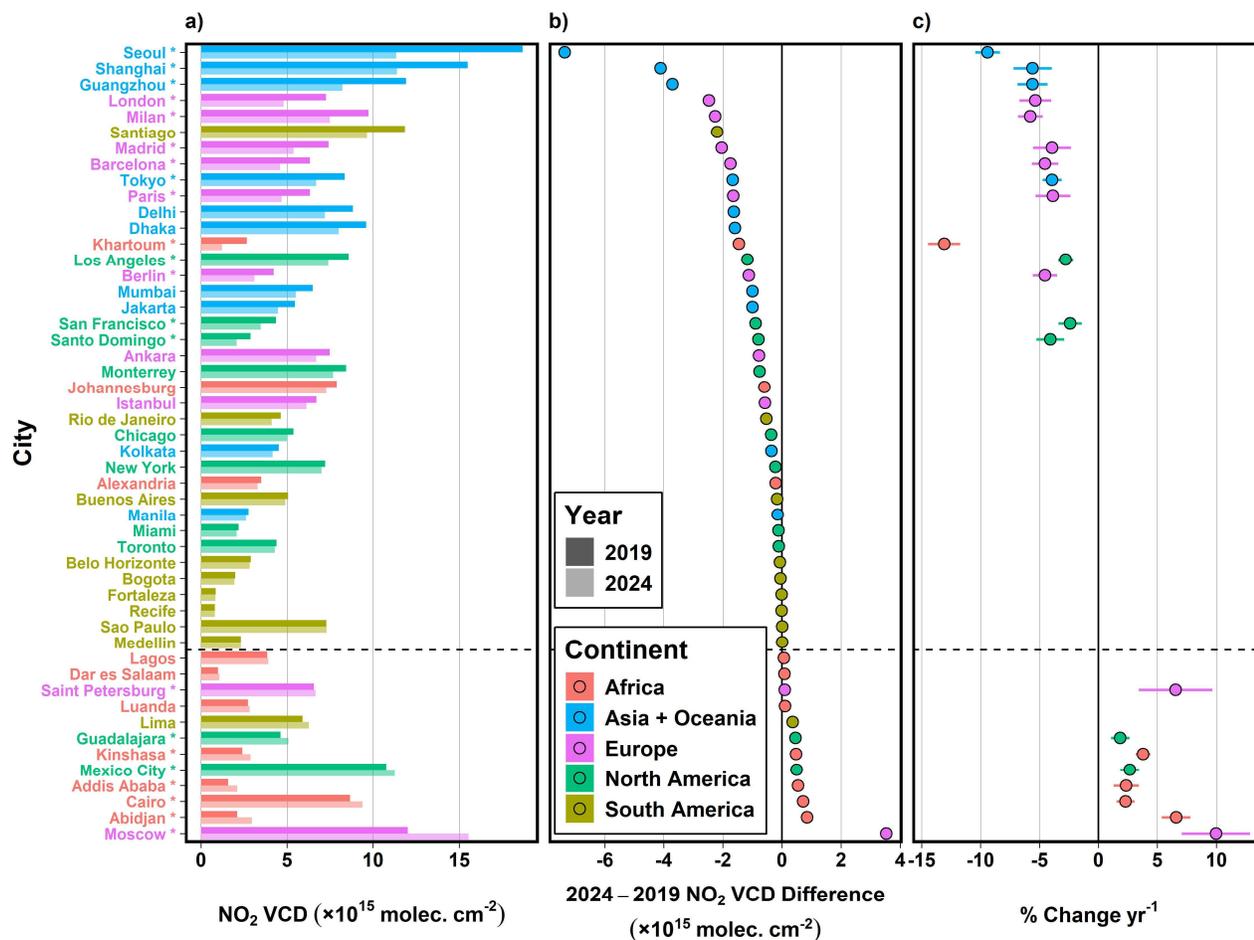
170 We use data from two inventories to evaluate NO_x emissions: (1) version 8.1 of the Emissions Database for Global
171 Atmospheric Research (EDGARv8.1; Crippa et al., 2024), and (2) the 2025 version of Community Emissions Data System
172 (CEDS; Hoesly et al., 2025). EDGAR provides annual summed total and sector-specific NO_x emissions at 0.1° × 0.1° spatial
173 resolution globally, derived using a bottom-up method that combines sector-level activity data with corresponding emissions
174 factors for energy generation, industrial sources, transportation, residential sources and agriculture, with data available through
175 2022. CEDS is a similar bottom-up inventory, also provided at 0.1° × 0.1° spatial resolution, but provides emissions estimates
176 at the monthly level through the end of 2023. Uncertainties are inherent in such emissions inventories, with a roughly 10-50%
177 uncertainty when aggregating emissions to the country level, and even larger uncertainty for individual grid points (Crippa et
178 al., 2018).

179 Like the handling of TROPOMI data (Sec. 2.3), we use GHS-SMOD to quantify annual NO_x emissions for each urban cluster.

180 **3 Global VCD Changes from 2019 to 2024 in Major Urban Areas**

181 Using the method outlined in Section 2.2.1, the GHS-SMOD urban cluster boundaries are used to determine mean TROPOMI
182 NO₂ concentrations for all urban clusters globally. Of all 11,534 GHS-SMOD urban clusters, 58.1% are in Asia and Oceania,
183 18.5% are in Africa, 10.9% are in Europe, 6.2% are in North America and 6.3% are in South America. Looking at VCD
184 changes from 2019 to 2024 in the 50 cities representing the ten most populous urban clusters on each continent, with Asia and
185 Oceania considered jointly, East Asian cities represent four and European cities represent five of the ten largest VCD decreases
186 (Fig. 2a). Seoul experienced the greatest absolute reduction in annual mean NO₂ VCD of any of these 50 cities (Fig. 2b),
187 representing a significant decrease of $-9.4 \pm 1.0\% \text{ yr}^{-1}$ ($p < 0.001$; Fig. 2c). London, England produced the greatest NO₂ VCD
188 decrease of the ten most populous European cities ($-5.4 \pm 1.3\% \text{ yr}^{-1}$; $p < 0.001$), occurring alongside the introduction of the
189 city's ultra-low emission zone introduced in 2019 and expanded in 2023, which has contributed to decreased local NO₂
190 concentrations (Hajmohammadi and Heydecker, 2022).

191 None of the ten largest South American cities experienced statistically significant changes in NO₂ VCD, with relative changes
192 typically less than $\pm 0.6 \times 10^{15} \text{ molecules cm}^{-2}$ (Fig. 2b). The most notable exception is Santiago, Chile, which experienced an
193 annual mean VCD difference of nearly $-2.2 \times 10^{15} \text{ molecules cm}^{-2}$ between 2019 and 2024. Of the largest North American cities,
194 significant decreases occurred in Los Angeles ($-2.8 \pm 0.6\% \text{ yr}^{-1}$; $p = 0.004$), and the San Francisco Bay Area ($-2.8 \pm 0.6\% \text{ yr}^{-1}$; p
195 $= 0.023$), while significant increases occurred in the Mexican cities of Guadalajara ($+1.9 \pm 0.8\% \text{ yr}^{-1}$; $p = 0.019$) and Mexico
196 City ($+2.7 \pm 0.8\% \text{ yr}^{-1}$; $p = 0.010$).



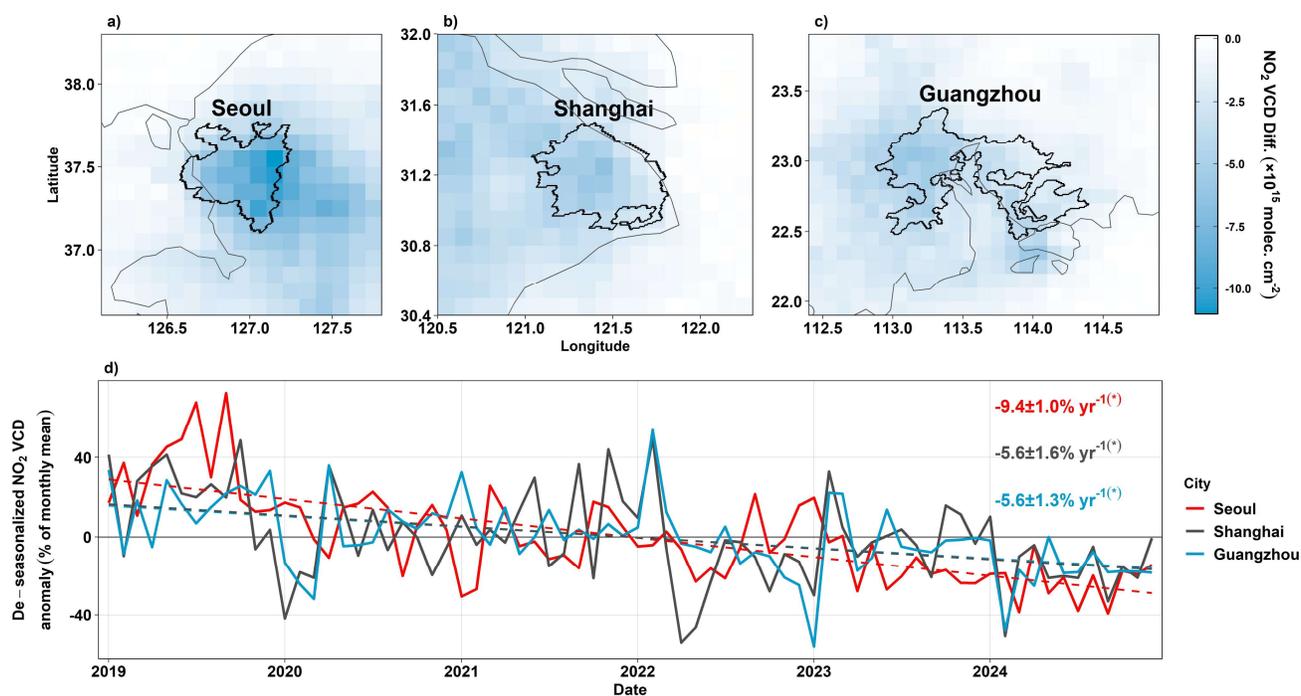
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199 **Figure 2: (a) NO₂ VCD in 2019 (dark bars) and 2024 (light bars) for the 10 most populous urban clusters on each continent, based**
 200 **on GHS-SMOD populations. (b) Absolute difference in NO₂ VCD for each city from 2019 to 2024. (c) NO₂ VCD percent change yr⁻¹**
 201 **from 2019 to 2024. Horizontal bars represent standard error, and colors correspond to the respective continent for each city. Cities**
 202 **are ordered by magnitude of absolute VCD decrease. Statistical significance is denoted with an asterisk by each city name. Only**
 203 **statistically significant results are reported in panel c.**

204 Most of the largest African cities experienced increased NO₂ VCDs from 2019 to 2024, with Abidjan, Ivory Coast experiencing
 205 the largest urban increase ($+6.6 \pm 1.2\%$ yr⁻¹; $p < 0.001$), with additional increases occurring in Cairo, Egypt ($+2.3 \pm 0.8\%$ yr⁻¹; p
 206 $= 0.006$); Addis Ababa, Ethiopia ($+2.4 \pm 1.1\%$ yr⁻¹; $p = 0.012$); and Kinshasa, DR Congo ($+3.8 \pm 0.6\%$ yr⁻¹; $p < 0.001$). In the
 207 Sudanese capital of Khartoum, NO₂ VCDs started decreasing in 2023, coinciding with the onset of conflict within Sudan (Guo
 208 et al., 2023; Fig. S10). This resulted in the largest absolute NO₂ VCD decrease of any African city from 2019 to 2024 (Fig.
 209 2b), and a decrease of $-13.1 \pm 1.4\%$ yr⁻¹ ($p < 0.001$).

210 Of the cities presented in Fig. 2, the three largest absolute decreases between 2019 and 2024 were in the East Asian cities of
 211 Seoul, South Korea (Fig. 3a); Shanghai, China (Fig. 3b); and Guangzhou, China (Fig. 3c). Decreases in Seoul coincide with
 212 known policies implemented by the South Korean government since the early 2000s to reduce local emissions, as well as
 213 changes in emissions that began following the COVID-19 pandemic (Ho et al., 2021; Seo et al. 2021). Moscow experienced
 214 the largest NO₂ VCD increase of any large GHS-SMOD city through 2024, with a VCD increase of +9.97% yr⁻¹ (p=0.001).
 215 This increase was accompanied by anomalously high monthly mean concentrations in early 2022 (Fig. S11), following the
 216 onset of the Russia-Ukraine war in Ukraine, when monthly mean NO₂ VCDs for March reached 59×10^{15} molecules cm⁻² (see
 217 Sec. 3.3).

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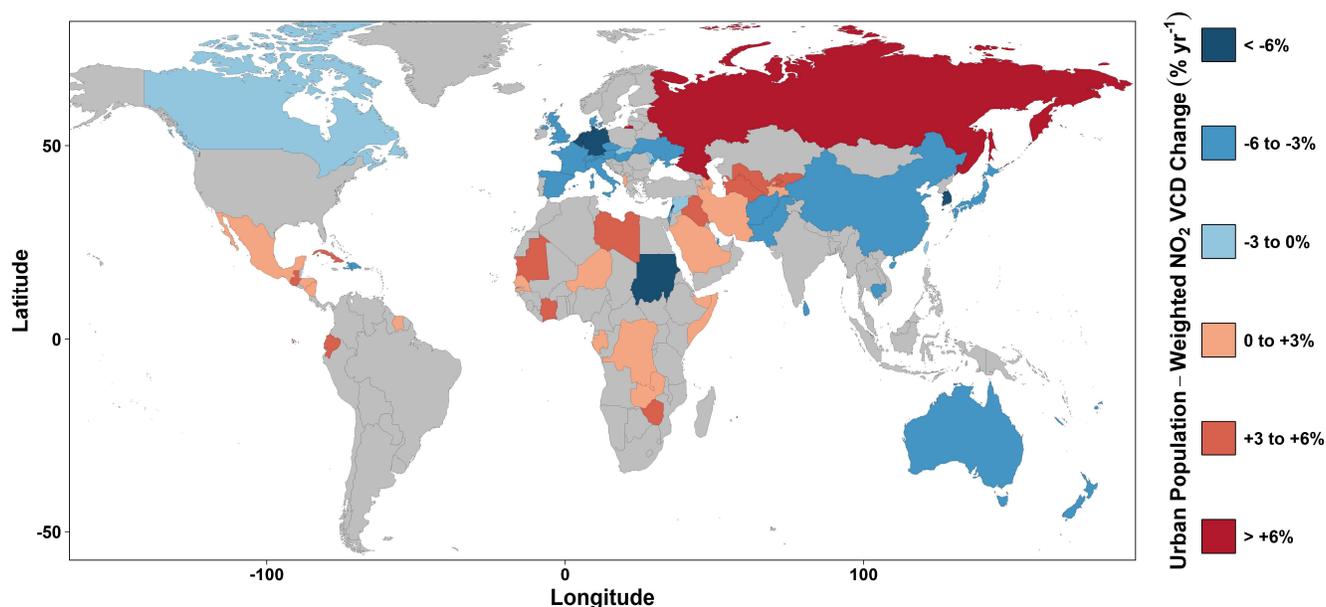
220 **Figure 3: Absolute change in mean annual NO₂ VCD from 2019 to 2024 for three East Asian cities: (a) Seoul, South Korea, (b)**
 221 **Shanghai, China and (c) Guangzhou, China. Colors in panels a-c show magnitude of VCD change, thin lines show national borders**
 222 **or coastlines, and thick lines show the GHS-SMOD urban boundary. (d) Solid lines show de-seasonalized monthly VCD anomaly**
 223 **from 01/2019 through 12/2024, colored by city. Dashed lines are produced from ordinary least-squares regression. The % change**
 224 **yr⁻¹, standard error and statistical significance is reported in the top right of panel d.**

225 4 Population-weighted Country-level Urban TROPOMI NO₂

226 Aggregating the NO₂ VCD changes to the country level by considering the population of each urban cluster (Eq. 1), we identify
 227 population-weighted VCD changes in countries globally (Fig. 4). The majority of urban NO₂ VCD increases were observed in
 228 much of Central America including Mexico, in Africa, in the Middle East and in Central Asia. Russia experienced the largest

229 population-weighted VCD increase of $6.2 \pm 3.6\% \text{ yr}^{-1}$ ($p = 0.046$). Broad urban VCD decreases were observed in numerous
230 countries across Western and Central Europe, as well as Eastern Asian countries. The largest urban population-weighted
231 decrease occurred in South Korea ($-8.74 \pm 0.9\% \text{ yr}^{-1}$; $p < 0.001$).

232



233

234 **Figure 4: Global spatial representation of the urban population-weighted NO₂ VCD % change yr⁻¹ from 2019 to 2024. Gray fill**
235 **denotes statistical insignificance ($p > 0.05$).**

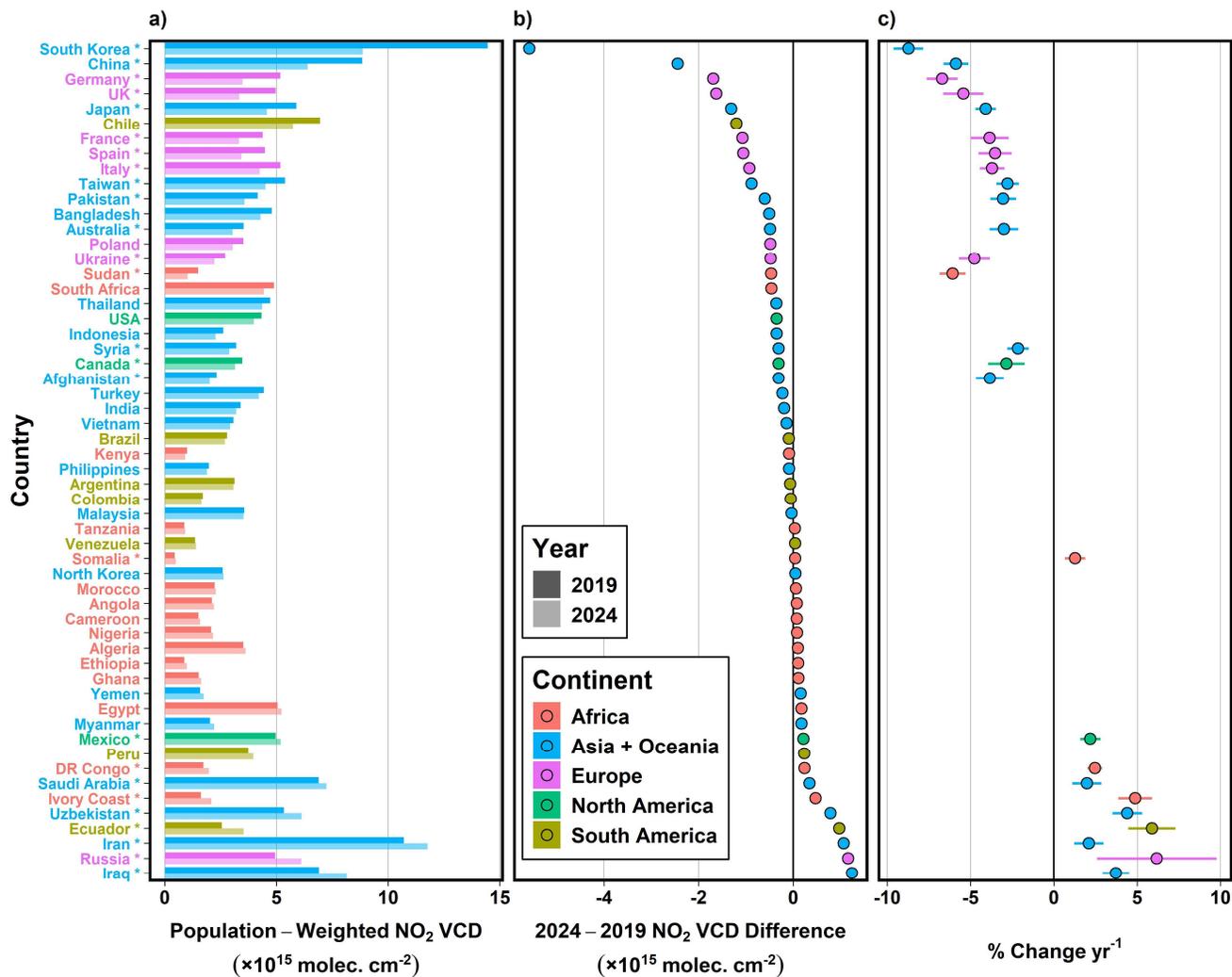
236 Much of the Middle East exhibited substantial increases in urban population-weighted NO₂ VCDs from 2019 to 2024, including
237 in Saudi Arabia ($+2.0 \pm 0.9\% \text{ yr}^{-1}$; $p = 0.009$), Iraq ($+3.7 \pm 0.8\% \text{ yr}^{-1}$; $p < 0.001$), and Iran ($+2.1 \pm 0.9\% \text{ yr}^{-1}$; $p = 0.013$), with broad
238 increases that extend beyond the urban environment. One of the most salient VCD decreases in the Middle East occurred in
239 Lebanon ($-8.5 \pm 1.0\% \text{ yr}^{-1}$; $p < 0.001$), coinciding with the country's severe economic and financial crisis that began in late 2019
240 (Harake et al., 2019). VCD decreases through 2024 were particularly stark in the Lebanese capital Beirut ($-7.9 \pm 1.1\% \text{ yr}^{-1}$;
241 $p < 0.001$). Additional Middle Eastern countries that exhibited decreased urban NO₂ VCDs through 2024 include much of Israel
242 ($-4.5 \pm 0.9\% \text{ yr}^{-1}$; $p < 0.001$), Qatar ($-3.4 \pm 1.2\% \text{ yr}^{-1}$; $p = 0.004$), and Afghanistan ($-3.8 \pm 0.8\% \text{ yr}^{-1}$; $p = 0.003$). Notable urban NO₂
243 VCD changes in less populated countries of Asia and Oceania include decreases in Cambodia ($-5.0 \pm 0.9\% \text{ yr}^{-1}$; $p < 0.001$), Sri
244 Lanka ($-5.4 \pm 0.9\% \text{ yr}^{-1}$; $p < 0.001$) and Australia ($-3.0 \pm 0.9\% \text{ yr}^{-1}$; $p = 0.008$). Urban increases were observed in much of Central
245 Asia, including Uzbekistan ($+4.4 \pm 0.9\% \text{ yr}^{-1}$; $p < 0.001$) and Turkmenistan ($+4.5 \pm 0.5\% \text{ yr}^{-1}$; $p < 0.001$).

246 NO₂ VCD decreases for more populous countries with an urban population of at least nine million were largest in East Asia,
247 including China ($-6.0 \pm 1.0\% \text{ yr}^{-1}$; $p < 0.001$) and Japan ($-4.1 \pm 0.6\% \text{ yr}^{-1}$; $p < 0.001$) (Fig. 5). Urban population-weighted VCD
248 decreases in South Korea were particularly pronounced, with a population-weighted concentration difference of -5.6×10^{15}
249 molecules cm⁻² between 2019 and 2024. In South Asia, the neighboring countries of Afghanistan ($-3.8 \pm 0.8\% \text{ yr}^{-1}$; $p = 0.003$)

250 and Pakistan ($-3.0 \pm 0.8\% \text{ yr}^{-1}$; $p=0.012$) exhibited some of the only significant country-level VCD decreases for the region.
251 Significant decreases also occurred in numerous countries of Western and Central Europe, with Germany experiencing the
252 largest VCD decrease in Europe through 2024 ($-6.7 \pm 0.9\% \text{ yr}^{-1}$; $p < 0.001$). Of the most-populous European countries, Russia
253 was the only country to experience increased population-weighted NO_2 VCDs through 2024.

254 A majority of larger African countries exhibited insignificant urban VCD changes, with 2024 population-weighted VCDs
255 changing by less than 0.25×10^{15} molecules cm^{-2} relative to 2019 levels (Fig. 5b). Exceptions include larger changes in Sudan
256 ($-6.1 \pm 0.8\% \text{ yr}^{-1}$; $p < 0.001$) and Ivory Coast ($+4.9 \pm 1.0\% \text{ yr}^{-1}$; $p < 0.001$). Middle Eastern and Central Asian countries
257 experienced some of the largest urban VCD increases, with Iraq experiencing the largest difference between 2019 and 2024
258 levels of any larger country ($+1.2 \times 10^{15}$ molecules cm^{-2}). Chile saw the largest difference in annual mean urban NO_2 VCD
259 between 2019 and 2024 of any South American country, due in large part to lower 2024 annual mean NO_2 VCDs in the capital
260 city of Santiago (Fig. 5b).

261



262

263 Figure 5: Same as Fig. 2 but presenting changes in country-level urban population-weighted NO₂ VCDs for countries with an urban
 264 population of at least nine million, based on urban cluster populations provided from GHS-SMOD.

265 **5 Regional TROPOMI NO₂ Vertical Column Densities from 2019 to 2024**

266 The following subsections describe NO₂ VCDs in five global subregions: Asia and Oceania, Africa, Europe, North America
 267 and South America

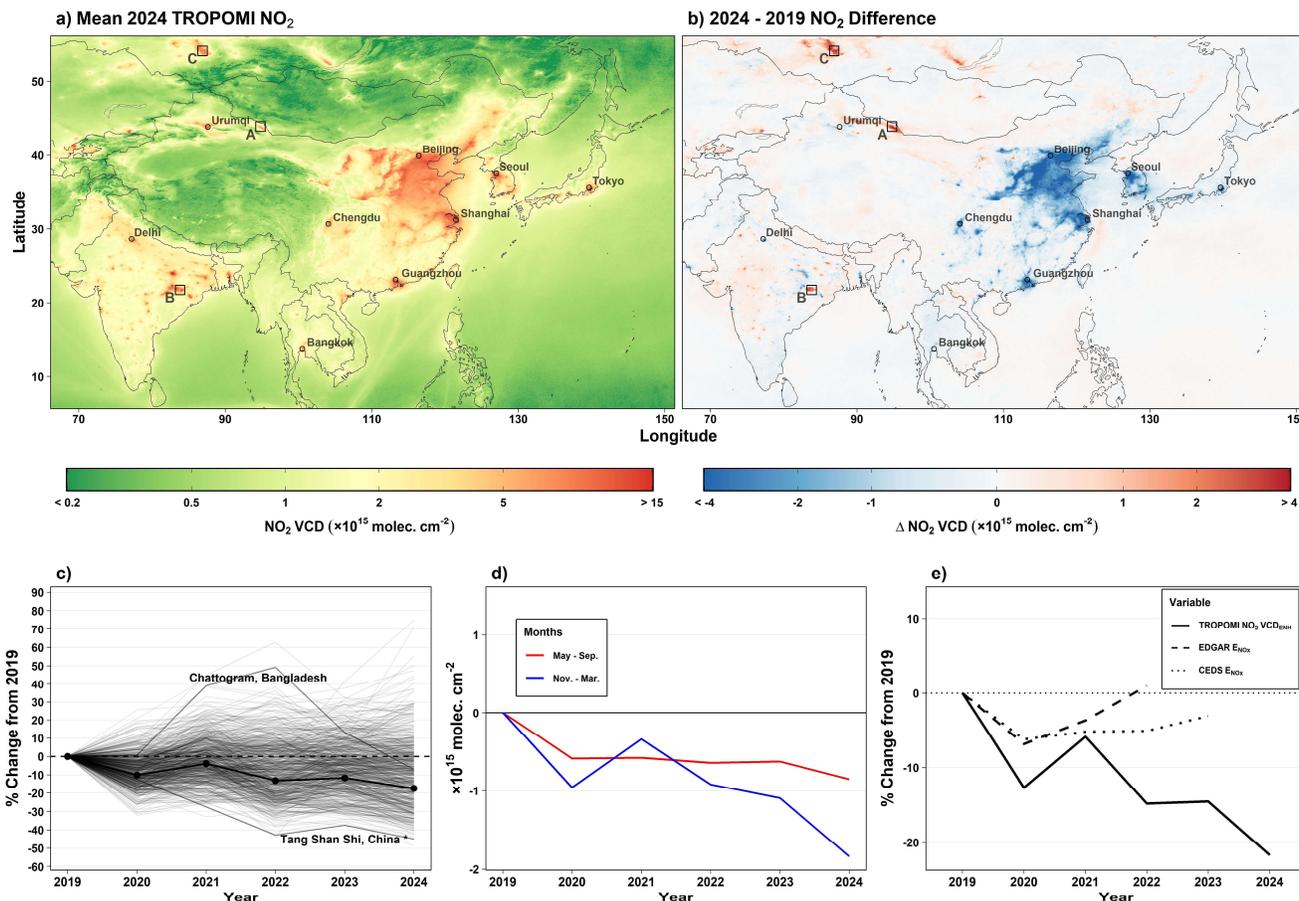
268 **5.1 Asia and Oceania**

269 North and East China, one of the most populated regions globally with approximately 11% of the 1000 largest GHS-SMOD
 270 cities, produced the broadest continuous expanse of 2024 annual mean NO₂ VCDs at or above 5×10^{15} molecules cm⁻² (Fig.

271 6a). Despite this, substantial VCD decreases were observed in this region from 2019 to 2024 (Fig. 6b). NO₂ concentrations
272 had already been decreasing in China prior to 2019 (Liu et al., 2016; de Foy et al., 2016), and the decrease continued after the
273 onset of the COVID-19 pandemic, during which numerous lockdowns throughout the country between 2020 and 2022 led to
274 reduced NO₂ concentrations (Zheng et al., 2021; Cooper et al., 2022; Levelt et al., 2022; Ma et al., 2023; Zhao et al., 2024).
275 The decrease in NO₂ also coincided with general Chinese government policies directed at reducing emissions, including stricter
276 emissions controls for industrial sources, energy generation and the transportation sector (Shi et al., 2022; Li et al., 2024).

277 In India, the largest differences in urban NO₂ VCD between 2019 and 2024 were observed in Delhi (-1.6×10^{15} molecules cm⁻
278 ²) and Mumbai (-1.0×10^{15} molecules cm⁻²), though neither city exhibited statistically significant decreases over that period.
279 Elevated NO₂ near numerous coal-fired power plants and coal mines is a common feature in India (Panda et al., 2023),
280 evidenced by the many apparent point sources in the 2024 annual average TROPOMI VCDs throughout the country (Fig. 6a).
281 NO₂ VCDs increased at many of these points sources from 2019 to 2024 (Fig. 6b), suggesting an increase in emissions from
282 energy production and use. In the Middle East and Central Asia, urban regions experienced some of the highest NO₂ VCDs
283 globally in the TROPOMI record (Fig. 7). The Iranian capital of Tehran by far has the largest annual average VCD in the
284 TROPOMI tropospheric NO₂ record for all GHS-SMOD cities, with annual mean values remaining above 30×10^{15} molecules
285 cm⁻² throughout the entirety of the TROPOMI record (Fig. S12).

286

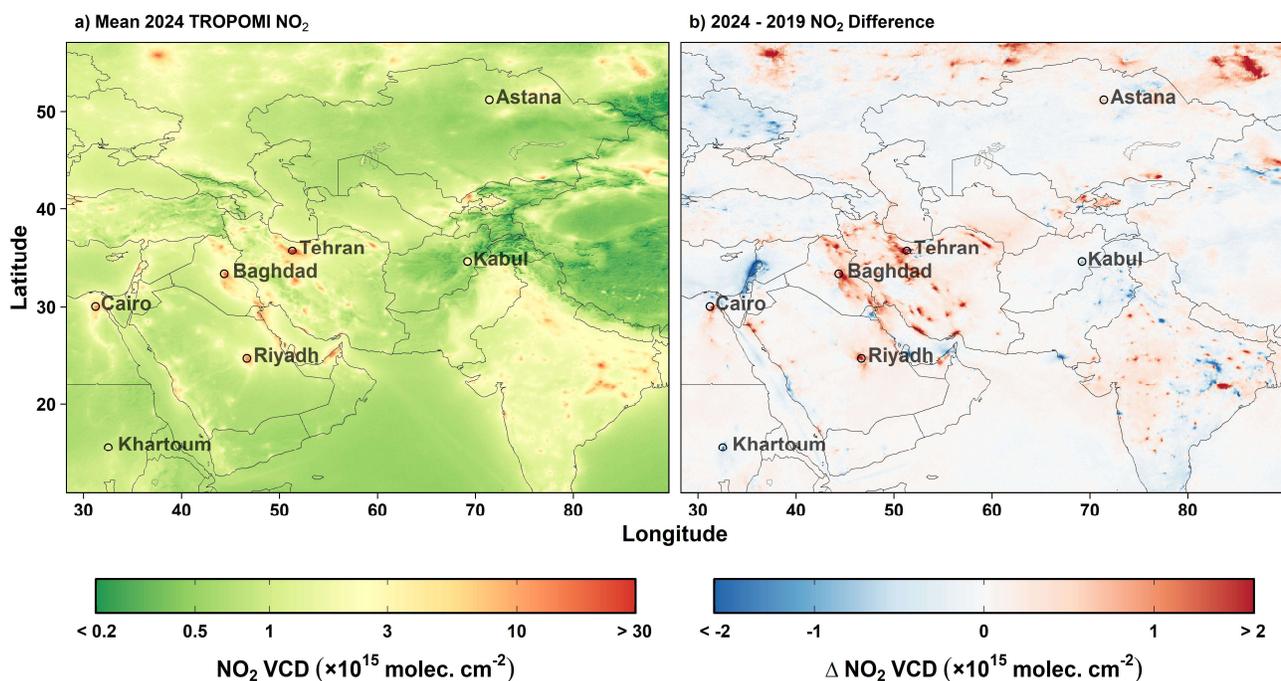


287

288 **Figure 6: (a) Mean 2024 TROPOMI NO₂ VCDs and (b) relative difference in annual mean TROPOMI VCDs between 2019 and**
 289 **2024, centered on South and East Asia. Regions A, B and C represent the Santanghu Basin, the Ib Valley and Kuzbass mining**
 290 **regions, respectively, as highlighted in Fig. 12. (c) Population-weighted percent difference in annual mean TROPOMI NO₂ VCD**
 291 **relative to 2019 levels for all GHS-SMOD urban clusters in Asia and Oceania (solid black line), and percent change in VCD for**
 292 **individual clusters with a population of at least 500,000 (gray lines). Asterisks denote statistical significance. (d) Absolute population-**
 293 **weighted difference in VCD for urban clusters in Asia and Oceania in May-September (red line) and November to March (blue line).**
 294 **(e) Relative difference in population-weighted TROPOMI NO₂ urban enhancement (VCD_{ENH}; solid line, 2019-2024), NO_x emissions**
 295 **from the EDGARv8.1 emissions inventory (dashed line, 2019-2022) and CEDS emissions inventory (dotted line, 2019-2023).**

296 Across Asia and Oceania as a whole, which contain a majority of all urban clusters globally, population-weighted NO₂ VCDs
 297 were approximately 17% lower in 2024 than in 2019 (Fig. 6c). One notable decrease in Asia occurred in the Chinese city of
 298 Tang Shan Shi, located to the east of Beijing, which experienced an NO₂ VCD decrease of nearly 45% from 2019 to 2024. The
 299 largest increase in Asia through 2024 occurred in the Mongolian capital of Ulaanbaatar, where the 2024 mean VCD was more
 300 than 70% larger than in 2019. Numerous Bangladeshi cities, including Chattogram, experienced substantially increased VCDs
 301 from 2020 through 2022, with VCDs decreasing again by 2024 to the near 2019 levels (Fig. S13).

302 Different seasons can have outside impact on the relative change in annual NO₂ VCD. In cities of Asia and Oceania, the bulk
 303 of the observed annual decreases through 2024 occurred during November – March (Fig. 6d), with a population-weighted
 304 decrease of -1.8×10^{15} molecules cm⁻². Although the absolute changes in November – March were larger than in May –
 305 September, the relative percent changes for the two periods were more comparable (Fig. S14).
 306



307
 308 **Figure 7: (a) Mean 2024 TROPOMI NO₂ VCDs and (b) relative difference in annual mean TROPOMI VCDs between 2019 and**
 309 **2024, centered on the Middle East and Central Asia.**

310 Urban NO₂ concentrations are not only influenced by local emissions, but also by advection of upwind pollutants into the urban
 311 boundary. We account for the role that upwind background concentrations may play in urban NO₂ concentrations by identifying
 312 changes in the urban enhancement of NO₂ (VCD_{ENH}), represented by the difference between NO₂ VCDs in the urban cluster
 313 and the urban background VCD. By removing the background concentrations, we expect that the percent change in VCD_{ENH}
 314 relative to a baseline year can be primarily attributed to changes in local, urban NO_x emissions. We then evaluate changes in
 315 VCD_{ENH} against changes in gridded NO_x emissions inventories from (1) the EDGARv8.1, with data available through 2022
 316 and (2) CEDS, with data available through 2023 (Fig. S15).

317 In Asia and Oceania, cities experienced sustained decreases in VCD_{ENH}, with population-weighted values 22.7% lower in 2024
 318 than in 2019 (Fig. 6e). Cities in Asia and Oceania experienced VCD_{ENH} that tracked relatively well with both inventories from
 319 2019 to 2021, with a mean difference of +4.0% (EDGARv8.1) and +3.6% (CEDS) between emissions and VCD_{ENH}. However,
 320 in 2022, EDGARv8.1 showed increased emissions and CEDS exhibited mostly unchanged emissions, while VCD_{ENH} exhibited

321 a sharp decrease for that year. This resulted in a percentage difference of +15.8% (EDGARv8.1) and +9.7% (CEDDS) between
322 emissions and VCD_{ENH} in 2022 relative to 2019 levels (Fig. 6e). The 2022 VCD_{ENH} decrease coincided with broad lockdowns
323 in China related to the COVID-19 pandemic, suggesting that EDGAR emissions may not reflect emissions decreases during
324 that lockdown period.

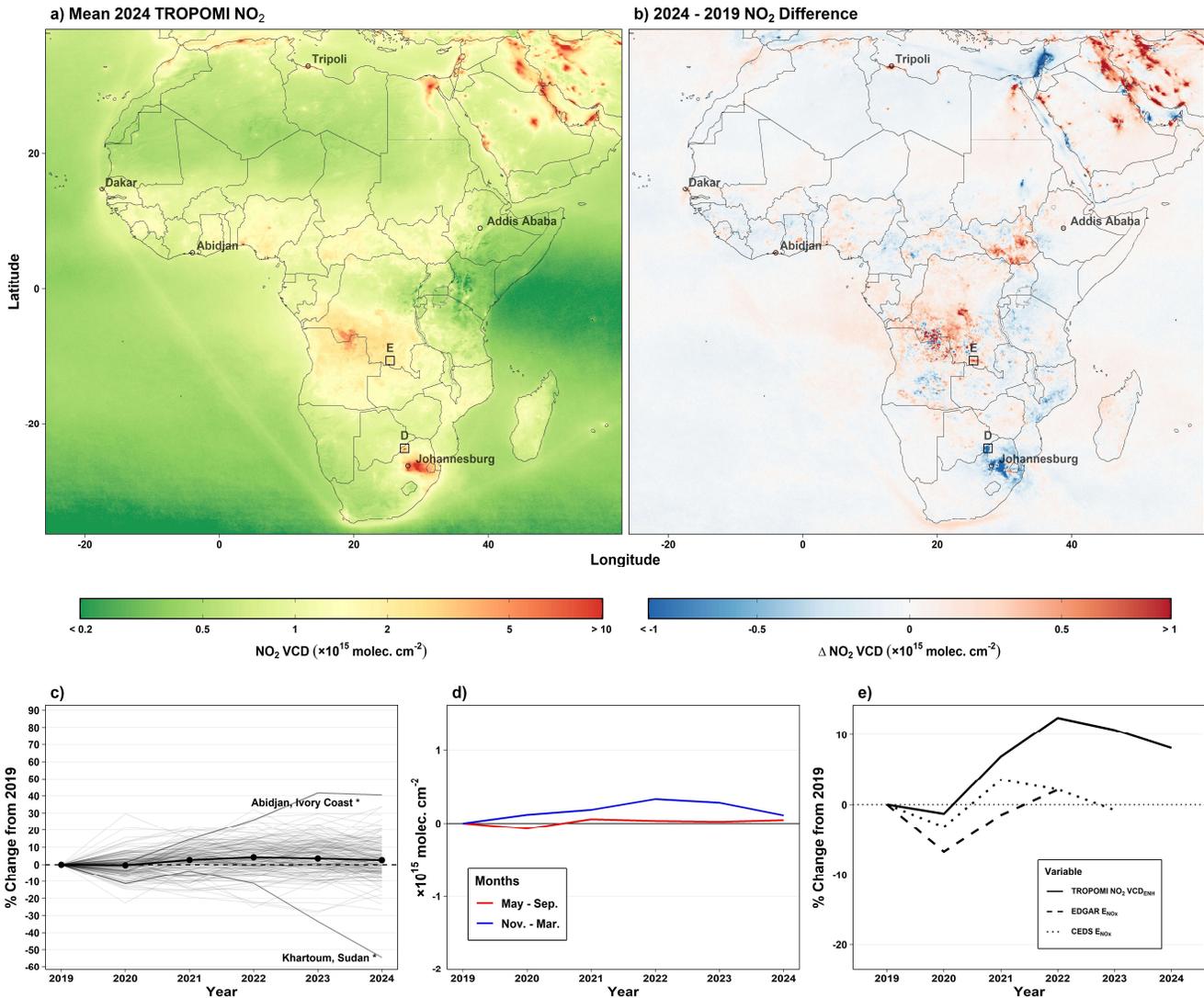
325 5.2 Africa

326 Areas to the east of Johannesburg, South Africa and the surrounding region exhibited the broadest enhanced NO_2 VCD for the
327 African continent in 2024 (Fig. 8a). Numerous surface coal mines and coal-fired power plants, particularly to the east of
328 Johannesburg, contribute to the region's NO_2 signature (Shikwambana et al., 2020). Cairo, Egypt represents the largest urban
329 NO_2 signature of any major urban region in Africa in 2024, when the annual mean NO_2 VCD reached 9.4×10^{15} molecules
330 cm^{-2} . From 2019 to 2024, Cairo experienced a statistically significant VCD increase of $2.3 \pm 0.8\% \text{ yr}^{-1}$ ($p = 0.006$). Along the
331 African Mediterranean coast, most urban areas showed increased NO_2 VCDs through 2024.

332 Through 2024, African cities experienced a gradual increase in population-weighted NO_2 VCD (Fig. 8c). The largest percent
333 increase occurred in Abidjan, the capital city of Ivory Coast, which experienced an increase in NO_2 VCD of more than 40%
334 from 2019 through 2024. Khartoum, Sudan experienced the largest percent decrease of any large African City, with mean 2024
335 levels nearly 60% lower than in 2019.

336 In African cities (Fig. 8d), population-weighted VCDs during November-March were 0.1×10^{15} molecules cm^{-2} larger in 2024
337 than 2019, with little to no change occurring on average during May – September. When evaluating changes in VCD_{ENH} in
338 African cities, population-weighted VCD_{ENH} were +8.1% larger in 2024 relative to 2019 levels (Fig. 8e). One distinct feature
339 for African cities is the lack of a pronounced decrease in VCD_{ENH} during 2020, coinciding with the onset of the COVID-19
340 pandemic, a feature observed on all other continents. Evaluating NO_x emissions inventories in African cities, we find a mean
341 difference of -8.0% (EDARv8.1) and -6.7% (CEDDS) between inventory NO_x emission and VCD_{ENH} changes, indicating a
342 potential underestimate in both emissions inventories in African cities for this period.

343

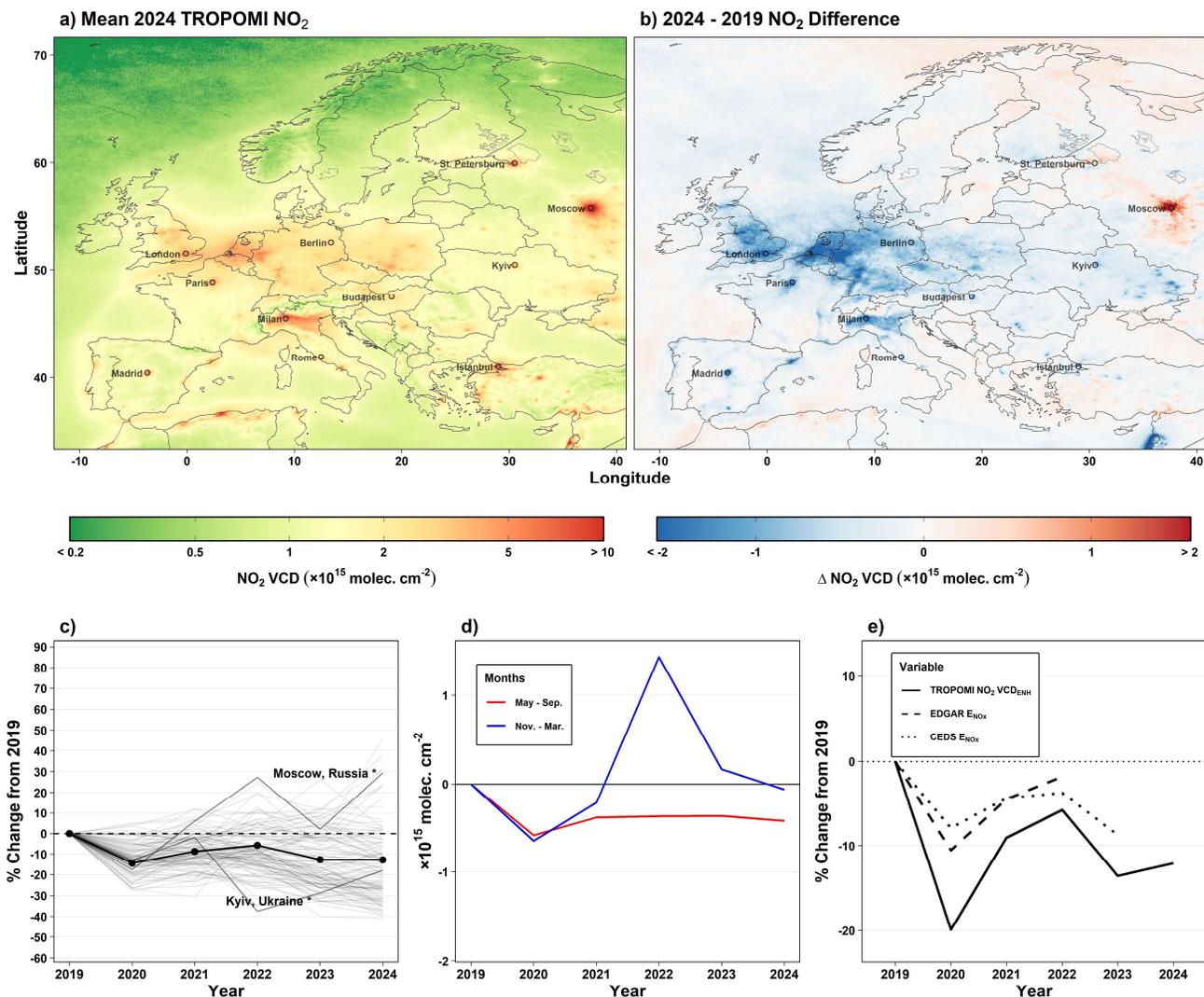


344

345 **Figure 8:** Same as Fig. 6, but for the African continent. Regions D and E in panels a and b represent the Grootegeluk and Kolwezi
 346 mines, respectively, as highlighted in Fig. 12.

347 **5.3 Europe**

348 NO₂ VCDs in Europe were largest in urban areas, with the largest 2024 mean VCD occurring in Moscow, Russia (15.5×10^{15}
 349 molecules cm⁻²) (Fig. 9a). Broad enhanced 2024 annual mean VCDs exceeding 4×10^{15} molecules cm⁻² were observed in a
 350 region encompassing Belgium, the Netherlands and western portions of Germany, with values exceeding 5×10^{15} molecules
 351 cm⁻² in the Po River Valley of northern Italy.



352
353 **Figure 9: Same as Fig. 6, but for Europe.**

354 Of the 1257 urban clusters in Europe, 1007 (80%) exhibited lower VCDs in 2024 than in 2019. Of the 53 European urban
 355 clusters with a population greater than 1,000,000, 2024 VCDs were lower than 2019 VCDs in 48 (91%), with the exception of
 356 Moscow and other cities of western Russia, which experienced increases (Fig. 9b). The broad decreases across large European
 357 cities are likely due to a combination of (1) a decrease in emissions that continued following the COVID-19 pandemic, (2)
 358 continued transition to alternative energy sources following the start of the Russia-Ukraine war in 2022 and (3) existing policies
 359 implemented within the EU (Matthias et al., 2021; Rokicki et al., 2023; Cifuentes-Faura, 2022). These policies include the

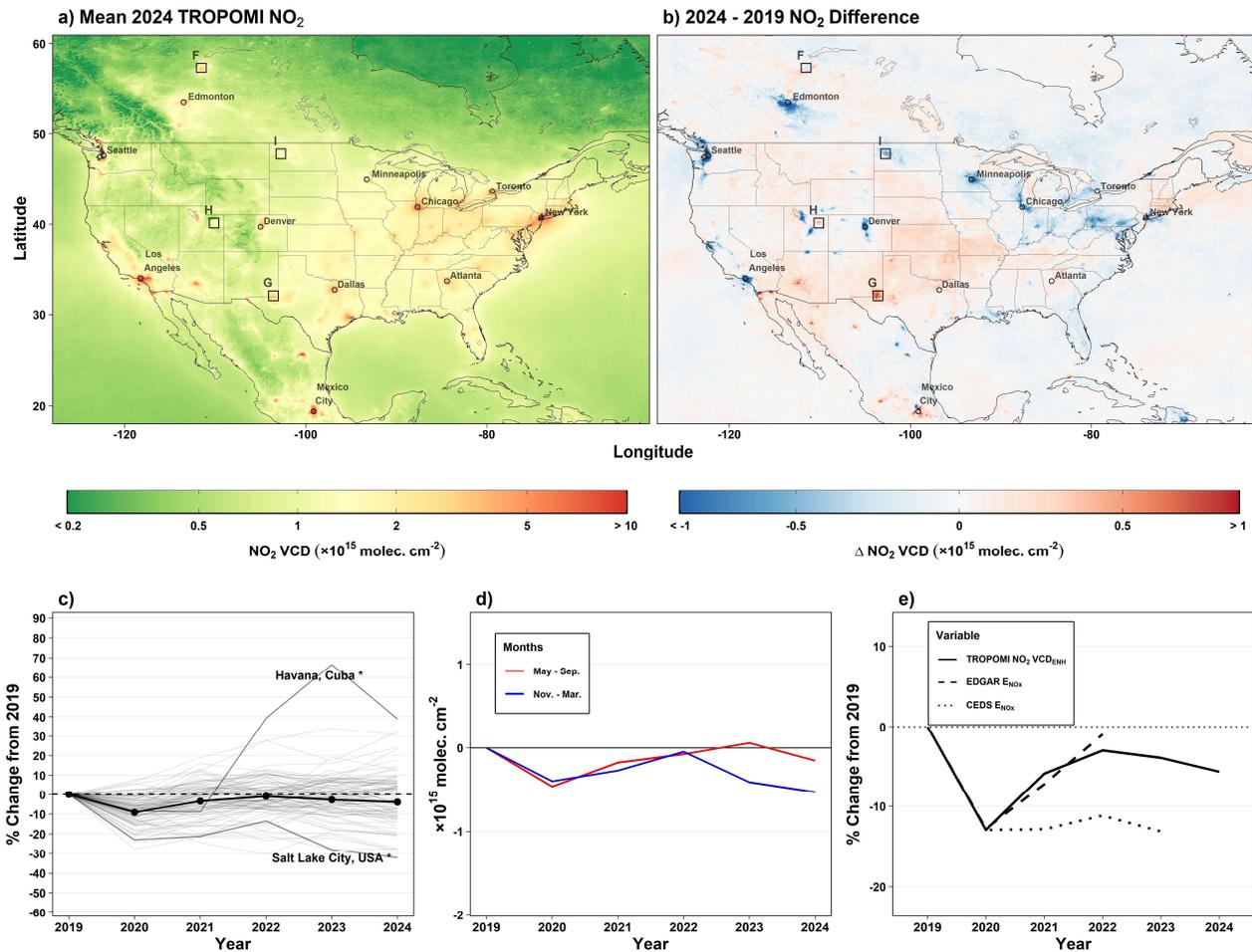
360 European Green Deal and European Climate Law, which promote zero-emission vehicles, stricter vehicle emissions targets
361 and updated industrial emissions regulations.

362 European cities experienced the most pronounced decrease in column NO_2 of any continent in 2020, with population-weighted
363 VCDs decreases by 16% from 2019 to 2020 (Fig. 9c). Previous work has attributed such decreases to the COVID-19 pandemic
364 (Cooper et al., 2022; Levelt et al., 2022). NO_2 VCDs rebounded marginally in 2021 and 2022, followed by decreases into 2023
365 and 2024. Decreases are more pronounced when only analyzing cities in the 27 member countries of the European Union (Fig.
366 S16). One notable feature within the European annual average VCDs is the contrasting VCD directionality in Russian and
367 Ukrainian cities in 2022, at the onset of the Russia-Ukraine War (Fig. S17). In the Ukrainian capital of Kyiv, annual VCDs
368 dropped nearly 40% in 2022 relative to 2019, coinciding with a large portion of the city fleeing due to conflict in and near the
369 city. To contrast this, VCDs increased nearly 30% in the Russian capital of Moscow during the same period. Following 2022,
370 VCDs in Kyiv increased steadily, while in Moscow, levels decreased in 2023 then increased again in 2024.

371 Population-weighted May – September VCDs decreased by 0.4×10^{15} molecules cm^{-2} (-10%) through 2024, while VCD
372 behavior during November – March has been less consistent, despite a sharp increase in winter-time levels in 2022 during the
373 onset of the Russia-Ukraine war (Fig. 9d). We note that the seasonal changes in Europe show more comparable winter and
374 summer changes if evaluating with Russian cities removed (Fig. S18). When accounting for background concentrations,
375 VCD_{ENH} in European cities experienced the largest drop in 2020 of any continent, with population-weighted VCD_{ENH}
376 decreasing by -20% from 2019 to 2020 (Fig. 9e). While both EDGARv8.1 and CEDS exhibited similar mean year to year
377 variability as VCD_{ENH} in European cities, changes in the inventories appeared underestimated, with each inventory estimate
378 exhibiting a mean percent difference relative to VCD_{ENH} of +6.0 and +5.9%, respectively. This suggests a slight underestimate
379 in emissions inventory decreases in European cities relative to observed VCD_{ENH} levels.

380 **5.4 North America**

381 Throughout North America, 2024 annual mean NO_2 VCDs were largest in urban regions, including Los Angeles (7.4×10^{15}
382 molecules cm^{-2}), New York (7.0×10^{15} molecules cm^{-2}), and Mexico City (11.3×10^{15} molecules cm^{-2}), as well as near fossil
383 fuel-fired power plant and mining operations (Fig. 10a). Most major cities in the U.S. and Canada exhibited decreased or
384 unchanged NO_2 VCDs (Fig. 10b). Phoenix, Arizona was one notable exception to these decreases, with mean 2024 VCDs 10%
385 higher than in 2019 (Fig. S19).



386

387 **Figure 10: Same as Fig. 6, but for North America. Regions F, G, H and I in panels a and b represent the Athabasca, Permian, Bakken**
 388 **and Uintah, respectively, as highlighted in Fig. 12.**

389 In Canada, the largest difference in VCD between 2024 and 2019 occurred in Alberta Province in and around Edmonton (-0.9
 390 $\times 10^{15}$ molecules cm^{-2} ; Fig. 10b), although decreases were not statistically significant for that period. In the U.S., aside from
 391 decreases in urban environments, the largest changes were observed in remote areas near coal power plants with reduced
 392 activity, e.g. near the decommissioned Navajo Generating Station in northern Arizona (Goldberg et al., 2021). Apparent within
 393 the U.S. is a slight increase in background concentrations in rural regions, particularly in the Central and Western U.S. It is
 394 unclear if this is due to an extension of the NO_2 lifetime due to decreasing VOCs and O_3 over this 6-year period (e.g., Laughner
 395 & Cohen 2019) or due to increased NO_x emissions in rural areas or both. Further work should investigate this.

396 In Mexico, Central America and the Caribbean, the largest VCDs are observed near Mexico City (11.3×10^{15} molecules cm^{-2})
 397 and Monterrey, Mexico (7.7×10^{15} molecules cm^{-2}), with numerous other notable urban signatures (Fig. 10a). The largest

398 urban increases were observed at sites in Northern Mexico, including Mexicali ($+6.1 \pm 0.9\%$ yr⁻¹; $p < 0.001$) and Hermosillo
399 ($+5.2 \pm 0.7\%$ yr⁻¹; $p < 0.001$). Additional notable changes occurred in the capital city of Santo Domingo, Dominican Republic
400 ($-4.1 \pm 1.2\%$ yr⁻¹; $p = 0.006$), and Havana, Cuba ($+11.2 \pm 1.7\%$ yr⁻¹; $p < 0.001$) (Fig. 10b).

401 Most North American cities experienced a decrease in annual NO₂ VCD of less than 10% in 2020, with concentrations
402 generally rebounding to 2019 levels by 2024 (Fig. 10c). Havana, Cuba was a notable exception of North American cities, with
403 VCDs increasing by nearly 70% through 2023 relative to 2019, with a slight decrease in 2024. Cities in the western U.S., such
404 as Salt Lake City and Denver experienced some of the largest percent decreases on the continent, decreasing by approximately
405 30% through 2024. The bulk of the observed annual decreases through 2024 in North American cities occurred during winter
406 (Fig. 10 d), with an average winter decrease of -0.5×10^{15} molecules cm⁻² during those months. In North America, VCD_{ENH}
407 decreased by 13% from 2019 to 2020 (Fig. 10e), compared with a decrease of 10% in overall urban VCD from 2019 to 2020,
408 and VCD_{ENH} remained approximately 7.5% below 2019 levels by 2024. Averaged for North America, population-weighted
409 EDGAR NO_x emissions and VCD_{ENH} exhibited a similar change relative to 2019 levels through 2022, with a mean difference
410 of +0.3%, while CEDS and VCD_{ENH} exhibited a larger mean difference of -6.1%, with differences most pronounced after 2020.
411 This suggests relatively good agreement between North American EDGAR and TROPOMI relative changes, while CEDS
412 emissions for the region may be underestimated from 2020 onward (Fig. 10e).

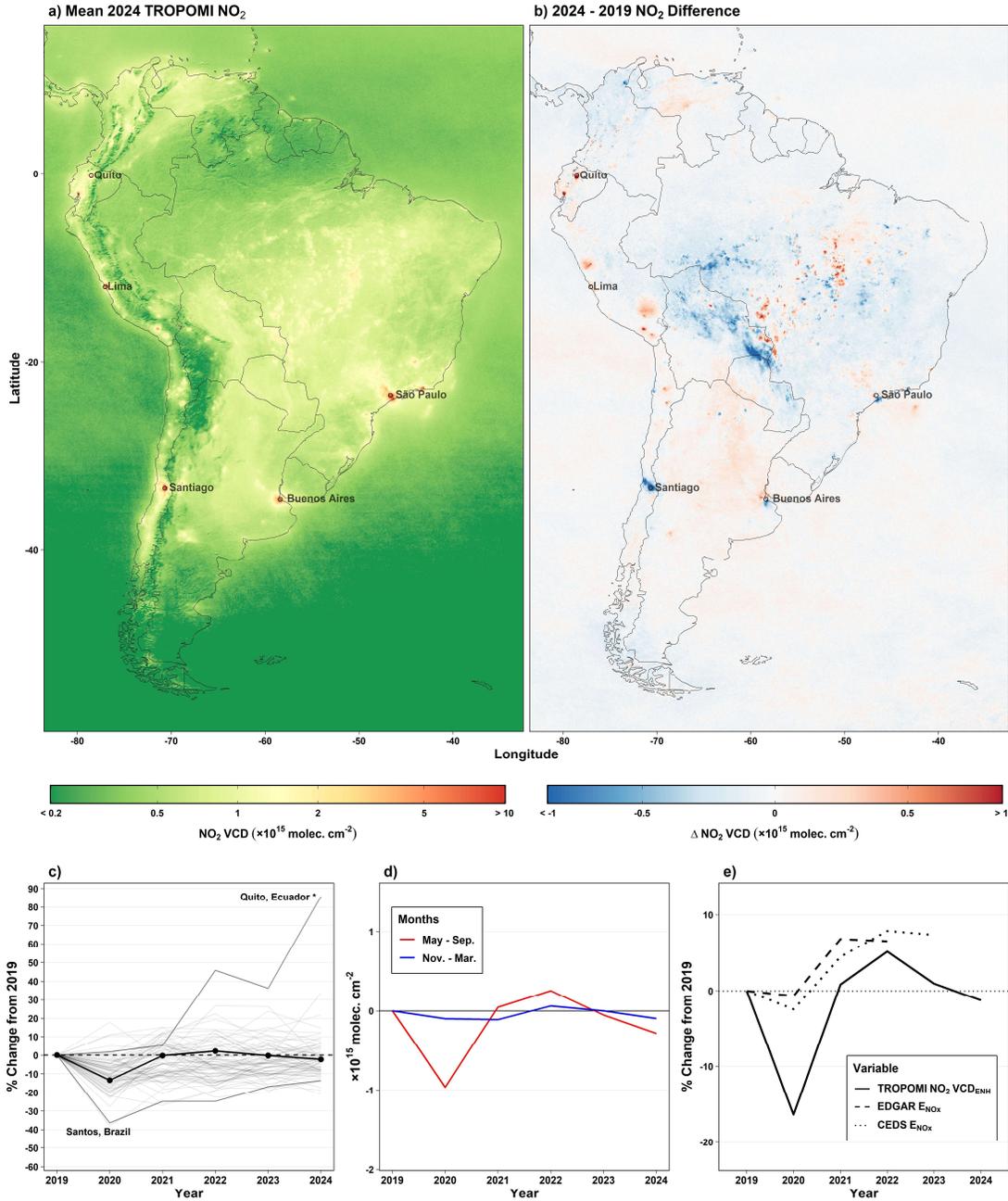
413 **5.5 South America**

414 The largest 2024 mean VCDs in South America are observed in urban regions, including near Lima, Peru (6.3×10^{15} molecules
415 cm⁻²); Santiago, Chile (9.7×10^{15} molecules cm⁻²); and Sao Paulo, Brazil (7.3×10^{15} molecules cm⁻²) (Fig. 11a). Regions near
416 Santiago experienced some of the largest differences in VCD in South America between 2019 and 2024 (Fig. 11b) (-2.2×10^{15}
417 molecules cm⁻²), while Quito, Ecuador experienced a significant increase for that period ($+12.7 \pm 1.9\%$ yr⁻¹; $p < 0.001$).

418 South American cities experienced a 10% population-weighted VCD decrease in 2020, with mean concentrations rebounding
419 to 2019 values by 2021 and remaining around those levels through 2024 (Fig. 11ce). One notable exception is Quito, Ecuador,
420 which experienced a VCD increase of over 85% through 2024. Santos, Brazil, an active port town southeast of São Paulo,
421 experienced one of the largest VCD decreases in South America, with a 35% decrease in VCDs from 2019 to 2020, followed
422 by sustained, gradual annual increases through 2024.

423 Seasonal changes impacted South American cities less than cities on other continents through 2024 (Fig. 11d), with mean
424 winter and summer VCDs both changing by less than 0.3×10^{15} molecules cm⁻² through 2024. Accounting for urban
425 background concentrations, South American cities experienced a population-weighted VCD_{ENH} decrease of 16% from 2019 to
426 2020, with concentrations rebounding to near 2019 levels by 2021 (Fig. 11e). Both EDGAR and CEDS estimated similar
427 relative population-weighted NO_x emission changes for the region, though neither inventory appeared to capture the robust
428 2020 decrease observed by TROPOMI (Fig. 11e). Both inventories experienced a similar mean difference between emissions

429 and VCD_{ENH} (+7.7% and +6.7%, respectively), suggesting that urban NO_x emissions in both inventories may be overestimated
 430 for the region.



431

432 **Figure 11: Same as Fig. 6, but for South America.**

433 6 TROPOMI NO₂ VCD Changes in Oil, Gas and Other Mining Regions

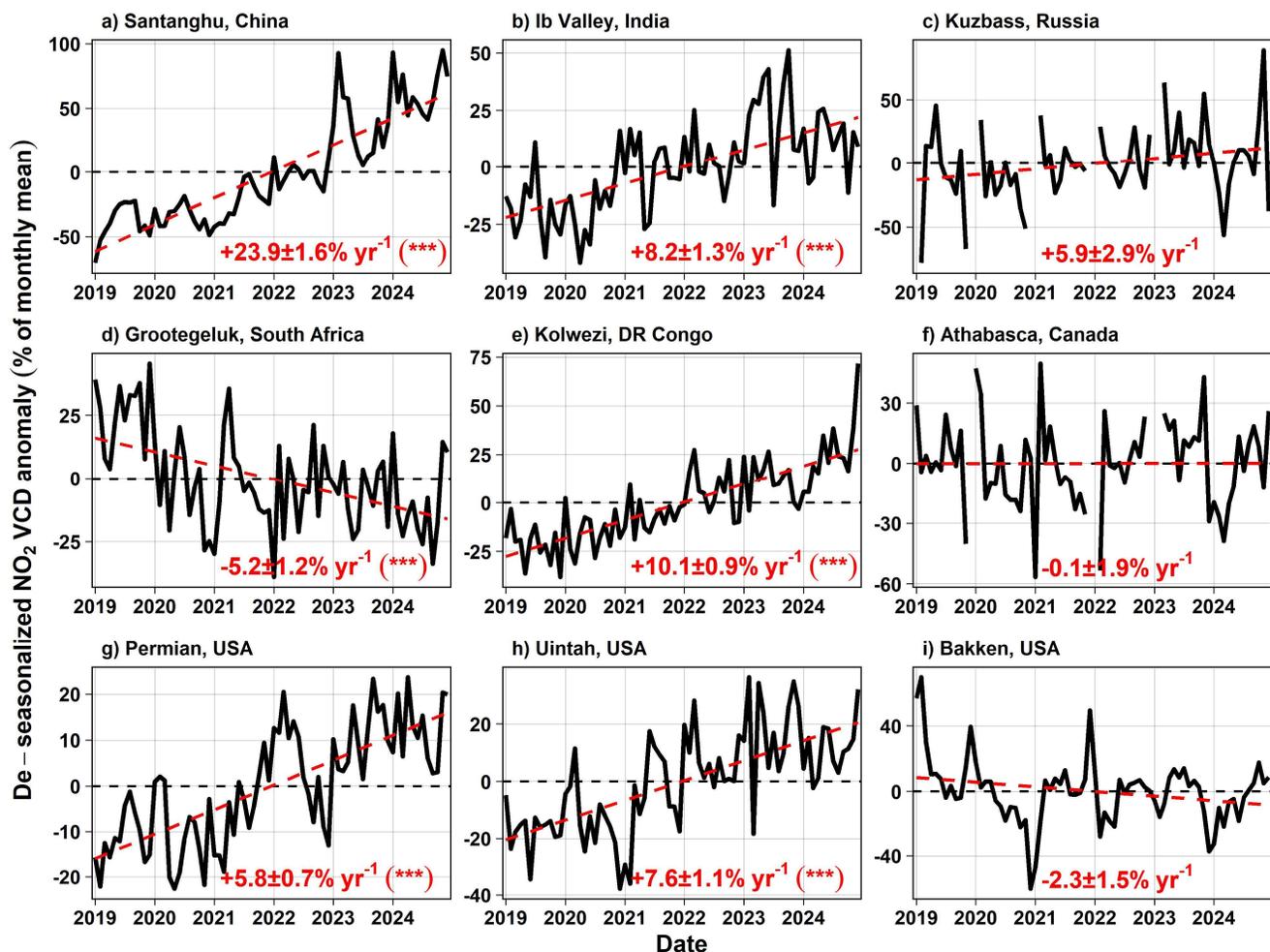
434 NO₂ can be readily observed over oil, gas, and other mining regions due to emissions from drilling and extraction equipment,
435 processing plants, compressors, truck traffic, and routine or episodic flaring. In these settings, increases or decreases in NO₂
436 can signify shifts in production levels or changes in mining activity. Because NO₂ responds quickly to changes in combustion-
437 related activity, satellite retrievals serve as an effective proxy for monitoring relative operational intensity in major extraction
438 regions (Dix et al., 2022).

439 Known coal-dominated mining regions showed pronounced NO₂ VCD increases from 2019 to 2024 (Fig. 12). The sparsely-
440 populated Santanghu Basin (Fig. 12a), a region in eastern Xinjiang Province with a relatively nascent coal mining industry
441 (Zhang et al., 2018; Liu et al., 2018), represented the most substantial increase in VCD over China through 2024 ($23.9 \pm 1.6\%$
442 yr^{-1} ; $p < 0.001$). The recent expansion of mining operations is evident in visible satellite imagery (Fig. S20). The largest regional
443 increase in VCD anywhere in India from 2019 to 2024 ($+2.1 \times 10^{15}$ molecules cm^{-2}) was observed in the Ib Valley in
444 northwestern Odisha state (Fig. 12b). The region contains multiple surface coal mines and coal-fired power plants (Varma et
445 al., 2015), with VCDs increasing at a rate of $8.2 \pm 1.3\% \text{ yr}^{-1}$ ($p < 0.001$). NO₂ VCDs near numerous other coal mines and power
446 plants throughout India exhibited changes, but NO₂ VCD increases were more prevalent than decreases. In the Kuzbass Region
447 of Siberia, one of Russia's largest coal mining regions, 2024 annual mean VCDs were 2.4×10^{15} molecules cm^{-2} higher than
448 in 2019, though annual changes were not statistically significant (Fig. 12c). A previous study identified a correlation between
449 space-based NO₂ observations and regional coal production in the Kuzbass region (Labzovskii et al., 2022), providing relevant
450 context for the observed VCD changes. Increased VCDs were also observed over rare earth metal mines. In a mining region
451 known as the Copperbelt in the south of the Democratic Republic of the Congo (DRC), broad NO₂ VCD increases were
452 observed, including at a large surface copper and cobalt mine near the city of Kolwezi (Fig. 12e). VCDs at the Kolwezi mine
453 increased at a rate of $10.1 \pm 0.9\% \text{ yr}^{-1}$ ($p < 0.001$) from 2019 to 2024. Numerous surface mines exist in the region, with most
454 observing increases in NO_x emissions from mining operations in recent years (Martínez-Alonso et al., 2023).

455 Not all coal regions experienced increased VCDs. Northwest of Johannesburg, South Africa in Limpopo Province, NO₂ VCDs
456 near the Grootegeluk surface coal mine, together with two adjacent power plants (Faure et al., 2010; Shikwambana et al., 2020)
457 decreased at a rate of $-5.2 \pm 1.2\% \text{ yr}^{-1}$ ($p < 0.001$) from 2019 to 2024 (Fig. 12d). The region represented one of the largest NO₂
458 signatures in Africa in 2024, despite the significant decrease for this period (Fig. 8a).

459 Oil and gas extraction areas in North America experienced diverse patterns. Annual mean NO₂ VCDs at the Athabasca oil
460 sands in Alberta, Canada were slightly lower in 2024 than in 2019, although the decrease for the period was insignificant ($p >$
461 0.05 ; Fig. 12f). The Bakken region in North Dakota, U.S. experienced a similarly insignificant decrease in VCDs (Fig. 12i).
462 Notable increases occurred in the Permian (Fig. 12g) and Uintah (Fig. 12h) Basins in the southwestern U.S. experiencing
463 significant increases of $5.8 \pm 0.7\% \text{ yr}^{-1}$ ($p < 0.001$) and $7.6 \pm 1.1\% \text{ yr}^{-1}$ ($p < 0.001$), respectively.

464



465

466 **Figure 12: Monthly time series of de-seasonalized NO₂ VCDs over selected oil, gas, and other mining regions. Black lines denote de-**
 467 **seasonalized VCDs, and dashed red lines represent ordinary least-squares regression for each site. Months with missing data lacked**
 468 **quality-assured TROPOMI observations. The % change yr⁻¹, standard error and statistical significance is reported each panel. Note**
 469 **the differing y-axis extents for each panel.**

470 7 Conclusions

471 We present a global analysis of urban TROPOMI tropospheric NO₂ VCD from 2019 to 2024 using GHS-SMOD-defined urban
 472 boundaries, encompassing more than 11,500 cities. Our results reveal statistically lower urban population-weighted NO₂ VCDs
 473 in 2024 than in 2019 in Asia and Oceania (-17%) and Europe (-13%) with particularly strong reductions in cities including
 474 Seoul (-9.4 ± 1.0% yr⁻¹; p < 0.001), Guangzhou (-5.6 ± 1.3% yr⁻¹; p < 0.001), and London, England (-5.4 ± 1.3% yr⁻¹; p < 0.001).
 475 These decreases generally reflect a combination of long-term emissions control policies and economic incentives, indicating
 476 policies to tackle NO₂ pollution have broadly worked. COVID-19 induced reductions in activity often caused a temporary NO₂
 477 reduction but is unlikely to have caused much of the long-term changes between 2019 and 2024. Conversely, urban NO₂ in

478 numerous African cities have increased over the same period, with Abidjan ($+6.6 \pm 1.2\% \text{ yr}^{-1}$; $p < 0.001$), Cairo ($+2.3 \pm 0.8\% \text{ yr}^{-1}$; $p = 0.006$) and Addis Ababa ($+2.4 \pm 1.1\% \text{ yr}^{-1}$; $p = 0.012$) representing larger cities that are leading the continent's upward
479 tendency. Though numerous populous North American cities exhibited significant VCD decreases, population-weighted urban
480 levels for the continent as a whole did not show a significant change. Similarly, South American cities exhibited an insignificant
481 VCD change from 2019 to 2024, apart from May-September in 2020. Population-weighted NO_2 VCDs increases were most
482 notable in countries in the Middle East and Africa, highlighting a potential degradation in air quality in regions of the world
483 that lack extensive ground-level monitoring.

485 Evaluating annual changes in TROPOMI NO_2 urban enhancements (VCD_{ENH})—the difference between mean urban and
486 background VCDs—against changes in EDGAR and CEDS NO_x emissions inventories, we highlight potential discrepancies
487 in inventory estimates in urban regions. In African, Asian and European cities, changes in VCD_{ENH} tend to exceed changes in
488 both EDGAR and CEDS emissions, pointing to potential inventory overestimates in NO_x emissions. In North America,
489 EDGAR agrees well with VCD_{ENH} (mean difference of 0.3% relative to 2019 values), while CEDS NO_x emissions are 6.1%
490 lower than VCD_{ENH} , relative to their respective 2019 values. These mismatches may stem from rapidly evolving emission
491 sources or limitations in the EDGAR and CEDS bottom-up inventory methods. Similar discrepancies in emissions inventories
492 in the Global South have been reported in previous studies (Ahn et al., 2023), suggesting larger emissions uncertainties in
493 regions where unmonitored emissions activity may be significant.

494 In most regions, VCD changes from 2019 to 2024 were driven by changes during the colder months (November – March).
495 This was most pronounced in Asian cities, where mean cold season VCDs decreased by $-1.2 \times 10^{15} \text{ molecules cm}^{-2}$ (-18%)
496 from 2019 to 2024, compared with warm season VCD decreases of $-0.5 \times 10^{15} \text{ molecules cm}^{-2}$ (-13%). Large changes in NO_2
497 were not confined to urban regions alone. We identified localized increases near fossil fuel and other mining operations,
498 including in the Santanghu Basin in China ($+23.9 \pm 1.6\% \text{ yr}^{-1}$; $p < 0.001$), the Permian ($+5.8 \pm 0.7\% \text{ yr}^{-1}$; $p < 0.001$) and Uintah
499 ($+7.6 \pm 1.1\% \text{ yr}^{-1}$; $p < 0.001$) Basins in the U.S., and the Copperbelt region of the DRC ($10.1 \pm 0.9\% \text{ yr}^{-1}$; $p < 0.001$), signaling
500 expanding industrial activity. In Khartoum and Kyiv, conflict and displacement drove sharp reductions in NO_2 , demonstrating
501 the utility of satellite data in detecting societal disruptions.

502 Several limitations of this work should be noted. First, satellite NO_2 column densities may not always reflect surface-level NO_2
503 concentrations, particularly in regions with vertically elevated sources. In urban areas dominated by surface-based
504 transportation emissions, NO_2 VCDs are likely more representative of surface exposure. However, in areas with tall-stack
505 sources, such as power plants, NO_2 columns may be decoupled from near-surface levels (Brett et al., 2025). Second, we assume
506 static city boundaries defined by the 2023 version of GHS-SMOD, with population estimates from 2020. This is likely a
507 reasonable approximation for urbanized regions in Europe and North America, where built-up area changes are slow, but may
508 introduce uncertainty in rapidly urbanizing regions of Africa and Asia over a six-year period. Future analyses could incorporate
509 time-varying urban boundaries to address this. Additionally, while many of the changes presented here reflect variability in
510 anthropogenic NO_x emissions, it is important to recognize that atmospheric chemistry also influences the observed NO_2

511 variability. Seasonal differences in photochemical lifetimes (i.e., longest in winter), boundary layer mixing (i.e., more vertical
512 mixing in summer), chemical partitioning between NO and NO₂ (i.e., the fraction of NO₂ is largest in winter), meteorological
513 variability, and contributions from additional emissions sources including soil NO_x and fire emissions, can all modulate the
514 magnitude and timing of observed NO₂ concentrations. These processes likely contribute to some of the regional and seasonal
515 differences highlighted in this study.

516 Taken together, these results demonstrate the utility of high-resolution satellite instruments for characterizing both broad
517 regional NO₂ signals and localized changes, and linking with anthropogenically induced factors such as urban growth,
518 industrial expansion, policy interventions, and conflict. This highlights potential in using TROPOMI observations as an
519 accountability agent to determine how local changes in human activities affect local and global air pollution. As the TROPOMI
520 record lengthens and newer, geostationary satellites come online and begin to detect changes in atmospheric composition,
521 continued space-based monitoring will be essential for improving our understanding of atmospheric composition and chemistry
522 around the globe.

523 **Data Availability.**

524 The level 3 annual and monthly average TROPOMI NO₂ VCDs are available at 10.5067/ACADNS5UBWPQ and
525 <https://doi.org/10.5067/KKPPL39PEIGE>, respectively. The GHS-SMOD urban boundaries can be downloaded from
526 <https://human-settlement.emergency.copernicus.eu/download.php?ds=smod>. The EDGARv8.1 NO_x emissions can be
527 downloaded from https://edgar.jrc.ec.europa.eu/dataset_ap81. The CEDS NO_x emissions can be downloaded from
528 <https://aims2.llnl.gov/>. Annual and monthly mean TROPOMI NO₂ VCDs for each GHS-SMOD urban cluster can be found at
529 <https://doi.org/10.5281/zenodo.18665781>.

530 **Supplement.**

531 The supplement contains additional figures related to the study, including: S1 Background NO₂ sensitivity in Beijing. S2
532 Background NO₂ sensitivity in Los Angeles. S3 Background NO₂ sensitivity in London. S4 Background NO₂ sensitivity in
533 Moscow. S5 Annual background NO₂ changes by continent. S6 Relative NO₂ VCD_{ENH} changes by continent. S7 Background
534 NO₂ for adjacent cities. S8 GHS-SMOD urban clusters example. S9 Data disaggregation example. S10 Khartoum NO₂ time
535 series. S11 NO₂ increases in three global cities. S12 Annual mean NO₂ in Tehran, Iran. S13 Annual mean NO₂ VCDs for
536 Bangladeshi cities. S14 Seasonal relative NO₂ changes by continent. S15 Annual mean NO₂ changes in the European Union.
537 S16 Annual mean NO₂ changes in Russian and Ukrainian cities. S17 Seasonal NO₂ changes by continent, without Russia. S18
538 NO₂ increases in three U.S. cities. S19 Satellite view of surface mines.

539 **Author Contribution.**

540 D.H. and D.G. contributed to the project design. D.G. processed and provided the annually- and monthly-averaged NO₂ vertical
541 column densities. All authors edited the manuscript.

542 **Competing Interests.**

543 The authors declare that they have no conflict of interest.

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