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Tracking air pollution and CO₂ emissions in 13,189 urban areas worldwide using large geospatial datasets

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Air pollution and climate change are urgent global concerns, with urban areas contributing heavily to both air pollutants and greenhouse gas emissions. Here we calculate fine particulate matter, nitrogen dioxide, and ozone concentrations and fossil-fuel carbon dioxide emissions per capita in 13,189 urban areas worldwide from 2005 to 2019 and analyze correlations between trends for these pollutants, leveraging recently-developed global datasets. Globally, we found significant increases in ozone (+6%) and small, non-significant changes in fine particulate matter (+0%), nitrogen dioxide (-1%), and fossil-fuel carbon dioxide emissions (+4%). Also, over 50% of urban areas showed positive correlations for all pollutant pairs, though results varied by global region. High-income countries with strong mitigation policies experienced decreases in all pollutants, while regions with rapid economic growth had overall increases. This study shows the impacts of urban environmental initiatives in different regions and provides insights for reducing air pollution and carbon dioxide emissions simultaneously.

With rapid economic and industrial growth across the globe, environmental pollution, including ambient air pollution and greenhouse gases (GHGs), has emerged as one of the most urgent global concerns. Exposure to air pollution is associated with adverse health effects such as morbidity and mortality due to cardiopulmonary diseases and cancer, as well as adverse birth outcomes¹⁻⁵. Ambient particulate matter and ozone were the fourth and 30th leading risk factors for premature deaths worldwide, responsible for ~5.2 million premature deaths in 20216, likely a substantial underestimate as only a subset of air pollutants and health endpoints were included. Also, the Intergovernmental Panel on Climate Change Sixth Assessment Report assessed that global surface temperature increased by 1.09 °C (90% uncertainty interval: 0.95-1.20) in 2011-2020 compared to 1850–19007. Climate change has resulted in substantial adverse impacts and irreversible losses to both human health and the environment, for example, through heightened occurrences of extreme weather events, disruption of human systems, diminished water and food security, and declines in biodiversity of ecosystems^{7,8}.

Urban areas play a pivotal role in emitting air pollutants and GHGs, especially areas experiencing intense urbanization without appropriate emission controls. Urban areas contribute over 70% of global GHG emissions due to dense, anthropogenic pollutant-emitting activities⁹. Similarly,

urban areas typically have higher levels of air pollution than non-urban areas, driven by increased polluting sources and their high density^{10,11}. Also, urban areas host large populations affected by diseases associated with air pollution. Over half of the global population resides in urban areas, a proportion expected to exceed two-thirds by 2050¹². Increased air pollution, combined with the larger number of people exposed, results in greater burdens of diseases attributable to air pollution in urban areas^{13,14}.

Substantial efforts have been undertaken to address environmental challenges globally. For example, the United Nations (UN) established multiple Sustainable Development Goals (SDGs) that focus on creating sustainable cities, expanding clean energy, and fostering climate action¹⁵. Additionally, the World Health Organization (WHO) has set air quality guidelines (AQGs) for key air pollutants to help countries achieve safe air quality levels and protect public health. On a city level, various mayoral networks, such as the C40 Cities Climate Leadership Group network (C40 Cities) and the International Council for Local Environmental Initiatives–Local Governments for Sustainability, are actively addressing climate change and environmental challenges. C40 Cities, for instance, has launched a Clean Air Accelerator, through which nearly 50 cities have set ambitious air pollutant emission reduction targets and are taking action¹⁶. Tracking progress towards these goals and guidelines is crucial for

¹Department of Environmental and Occupational Health, Milken Institute School of Public Health, George Washington University, Washington, DC, USA. ²Department of Energy, Environmental & Chemical Engineering, Washington University in St. Louis, St. Louis, MO, USA. ³Department of Environmental Sciences and Engineering, Gillings School of Global Public Health, University of North Carolina, Chapel Hill, NC, USA. —e-mail: sanenberg@email.gwu.edu accurately quantifying disease burdens related to climate change and air pollution, as well as assessing the societal benefits of actions taken to achieve progress.

Recent advances in satellite remote sensing and geospatial modeling methods have led to improvements in global gridded pollution datasets, which allow for more accurate and consistent estimation of urban air pollution and GHG emissions worldwide^{13,17-21}. For example, in 2022, three studies estimated air pollutant concentrations and associated disease burdens in ~13,000 cities globally, each for one of three ubiquitous air pollutants: fine particulate matter with an aerodynamic diameter ≤2.5 µm $(PM_{2.5})$, nitrogen dioxide (NO_2) , and ozone $(O_3)^{13,20,21}$. While these studies provided comprehensive urban estimates for each pollutant individually, exploring relationships between temporal trends among these pollutants and fossil-fuel carbon dioxide (FFCO₂) emissions can reveal additional insights. Because air pollutants and GHGs are largely co-emitted from fossil fuel and biofuel combustion²², positive or negative correlations among air pollutants and GHGs, hereafter collectively referred to as "pollutants", can suggest drivers of these trends. For example, rising FFCO2 coupled with declining NO2 might indicate that end-of-pipe emission controls are reducing NO2 while carbon-based fuel combustion is continuing to emit FFCO₂ unabated²². Declining FFCO₂ and NO₂ might indicate that fossil fuel combustion itself is declining-either through population decreases, increased energy efficiency, or transitions to renewable energy and electrification²². Conversely, increasing FFCO₂ and NO₂ simultaneously could indicate that emission sources are growing. Also, trends in these pollutants are influenced to varying degrees by natural sources (e.g., wildland fires, windblown dust), which are also increasingly linked to climate change and human activities in the long term²³. However, most studies investigating relationships among these pollutants are based on emission scenarios rather than historical observations^{24–26}, limiting our ability to analyze these relationships in real-world conditions.

This study aims to develop a multi-pollutant database for all urban areas worldwide and to analyze temporal trends of the four pollutants during 2005–2019, where datasets for each pollutant are available and were not influenced by the COVID-19 pandemic. We firstly calculate population-weighted average concentrations of PM_{2.5}, NO₂, and O₃, as well as FFCO₂ emissions per capita across 13,189 urban areas, leveraging recently-developed global gridded datasets of these pollutants and applying consistent methods to enable inter-city comparisons, and then test the correlations between temporal trends of these pollutants. Additionally, since governmental and non-governmental actors use different approaches for defining urban areas, we explore the influence of urban boundary definitions on urban pollution estimates and their temporal trends.

Results

Global distributions in 2019

In 2019, the global mean of population-weighted annual average $PM_{2.5}$ concentrations across all urban areas was 37.7 $\mu g/m^3$, which was 7.5 times larger than the WHO AQG of 5 $\mu g/m^3$ (Fig. 1). The global mean of urban NO $_2$ concentrations was 7.1 ppb, exceeding the WHO AQG (5.3 ppb ≈ 10 $\mu g/m^3$) by 34%. The global mean of urban O $_3$ concentrations (ozone season daily maximum 8 h mixing ratios (OSDMA8), see details in Methods) was 51.2 ppb, which was 67% higher than the WHO AQG (30.6 ppb ≈ 60 $\mu g/m^3$). The global median of FFCO $_2$ per capita across all urban areas was 2.8 metric tons in 2019.

We found distinct geographical patterns between urban $PM_{2.5}$, NO_2 , and O_3 concentrations and $FFCO_2$ per capita in 2019; nevertheless, some similar patterns emerged across pollutants with certain regions consistently standing out as hotspots (Fig. 1). Among all the cities, the highest levels of $PM_{2.5}$ were primarily observed in India (Indian cities mean: 61.5 vs. global mean: 37.7 μ g/m³), forming a highly polluted cluster in South Asia alongside cities in Bangladesh and Pakistan. Also, we observed clusters of urban areas with high $PM_{2.5}$ levels (> 90th percentile across all urban areas globally; 72.2 μ g/m³) in Nigeria, Saudi Arabia, and China. Urban areas with elevated NO_2 levels, exceeding the 90th percentile (12.1 ppb), were primarily in

China and High-income countries (based on the Global Burden of Disease (GBD) super-region categorization, see details in Supplementary Fig. 1 and Methods). For O₃, similarly to PM_{2.5}, cities in India had the highest levels, averaging 66.7 ppb which is 30% greater than the global mean (51.2 ppb). Urban areas in only five countries-India, Pakistan, Nepal, China, and Bangladesh-were observed to surpass the 90th percentile of O₃ concentrations (69.3 ppb), forming a highly-polluted cluster in South Asia and Southeast Asia, East Asian, and Oceania super-regions. Lastly, urban areas with FFCO₂ per capita levels greater than 90th percentile (19.0 metric tons) were heterogeneously distributed worldwide. The majority of cities above the 90th percentile were in China (25%), India (18%), and Iran (5%). When we compared spatial patterns across the pollutants, we found overall consistent geographical distributions of highly polluted cities, particularly between PM_{2.5} and O₃ concentrations. South Asia (largely due to India) and China commonly had relatively higher levels of PM_{2.5} and O₃ concentrations compared to other regions. Accordingly, in additional analysis, PM_{2.5} and O_3 showed the strongest positive correlation (r = 0.69 and p-value < 0.0001) among all the possible pairs of pollutants in 2019 (Supplementary Fig. 2). When testing correlations separately in each GBD super-region, while this analysis showed different results of correlations across GBD super-regions, all GBD super-regions showed positive correlations with statistical significance between PM_{2.5} and O₃, except for Latin America and Caribbean (Supplementary Fig. 2).

Temporal trends for 2005-2019

We also found different temporal trends across population-weighted annual average PM2,5, NO2, and OSDMA8 (O3) concentrations and FFCO₂ per capita (Fig. 2). Globally, urban PM_{2.5} concentrations showed an increase from $37.2 \,\mu\text{g/m}^3$ in 2005 to a peak of $41.4 \,\mu\text{g/m}^3$ in 2011, followed by a decline to 37.7 μg/m³ in 2019. Across GBD super-regions, South Asia showed the highest PM_{2.5} levels throughout the entire study period and even the largest increase from 51.5 µg/m³ in 2005 to 61.4 µg/ m³ in 2019. Central Europe, Eastern Europe, and Central Asia; Latin America and Caribbean; and High-income countries exhibited lower PM_{2,5} concentrations than the global mean over the entire study period with constant or decreasing trends. The global mean of urban NO2 concentrations showed little change across the study period, remaining within the range of 7.0-7.4 ppb. High-income countries; Southeast Asia, East Asia, and Oceania; and Central Europe, Eastern Europe, and Central Asia showed higher NO2 levels than the global mean. However, NO2 concentrations had steadily decreased in High-income countries and Central Europe, Eastern Europe, and Central Asia. The global mean of urban O₃ concentrations increased from 46.9 ppb in 2005 to 51.2 ppb in 2019. South Asia and Sub-Saharan Africa showed the greatest increases (54.6-66.3 ppb in South Asia and 40.1 to 46.2 ppb in Sub-Saharan Africa). Notably, South Asia maintained the highest O₃ concentrations among all the regions. The global median of urban FFCO2 per capita did not show large fluctuations from 2005 (2.56 metric tons) to 2019 (2.77 metric tons). High-income countries; Central Europe, Eastern Europe, and Central Asia; and North Africa and Middle East exhibited the highest levels of FFCO2 per capita over the study period. Among these regions, only High-income countries showed a consistent decrease from 6.23 metric tons in 2005 to 5.12 metric tons in 2019. Temporal trends in North Africa and Middle East experienced increases in FFCO₂ per capita until 2011 to decreasing trends afterward, resulting in an overall decline over the whole study period (2005: 4.60 metric tons, 2011: 5.06 metric tons, and 2019: 3.88 metric tons). In contrast, Central Europe, Eastern Europe, and Central Asia showed decreasing trends until 2014 to increasing trends afterward, which led to a small overall decrease (2005: 5.26 metric tons, 2014: 4.43 metric tons, and 2019: 5.09 metric tons). Southeast Asia, East Asia, and Oceania experienced a consistent increase from 2.61 metric tons in 2005 to 3.24 metric tons in 2019, starting to exceed the global median in 2010.

When considering the temporal trends of the four pollutants simultaneously as percent changes from the average values from 2005–2007 to

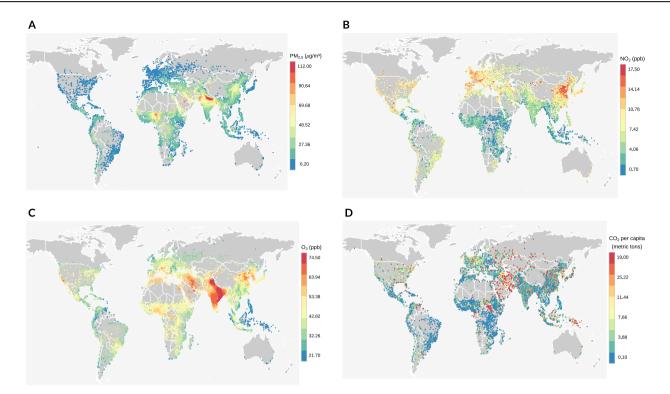


Fig. 1 | Global distributions of population-weighted annual average $PM_{2.5}$, NO_2 , and OSDMA8 (O₃) concentrations, and FFCO₂ per capita in urban areas in 2019. Values under the 1st percentile or above the 99th percentile (90th percentile for FFCO₂ per capita) were consolidated as 1st percentile or 99th/90th percentile values. A Population-weighted annual average $PM_{2.5}$ (μ g/m³), where n = 13,186, mean = 37.7 μ g/m³, 1st percentile = 6.2 μ g/m³, and 99th percentile = 112.0 μ g/m³. B Population-weighted annual average NO_2 (ppb), where n = 13,036, mean = 7.1

ppb, 1st percentile = 0.7 ppb, and 99th percentile = 17.5 ppb. C Population-weighted OSDMA8 (O₃) (ppb), where n = 12,942, mean = 51.2 ppb, 1st percentile = 21.7 ppb, and 99th percentile = 74.5 ppb. D FFCO₂ per capita (metric tons), where n = 13,185, median = 2.8 metric tons, 1st percentile = 0.1 metric tons, and 90th percentile = 19.0 metric tons. Dark gray areas indicate lands, light gray areas indicate oceans, and white lines indicate country boundaries.

those from 2017-2019, we found unique patterns varying by GBD superregion (Fig. 3). Regarding global trends, only urban O₃ concentration showed a statistically significant trend, an increase of 6%. Global urban PM_{2.5}, NO₂, and FFCO₂ per capita in 2017–2019 remained at levels similar to those in 2005–2007 (PM_{2.5}: +0%, NO₂: -1%, and FFCO₂ per capita: +4%), and these trends were not statistically significant. Across all the regions, only High-income countries showed declining trends in all four pollutants with statistical significance (PM_{2.5}: -19%, NO₂: -7%, O₃: -4%, and FFCO₂ per capita: -17%). Central Europe, Eastern Europe, and Central Asia showed significantly decreasing trends in three air pollutants (PM_{2.5}: -9%, NO₂: -5%, and O₃: -7%) but not in FFCO₂ per capita. In contrast, no decreasing trends were observed in Latin America and Caribbean; South Asia; and Sub-Saharan Africa. Latin America and Caribbean had nonsignificant/unchanging trends except for O_3 (+ 5%). South Asia showed the greatest increases with statistical significance in PM2.5 (+14%) and O3 (+18%) among all the regions. Sub-Saharan Africa showed simultaneous increases in NO_2 (+9%), O_3 (+9%), and FFCO₂ per capita (+8%). Regions with slowed population growth during the study period (Central Europe, Eastern Europe, and Central Asia and High-income countries) had overall decreasing trends, in contrast to regions with high-growing populations such as Latin America and Caribbean; South Asia; and Sub-Saharan Africa (Fig. 3).

The other two regions–North Africa and Middle East and Southeast Asia, East Asia, and Oceania–showed opposing trends between air pollutants and FFCO₂ per capita. North Africa and Middle East experienced increasing trends in air pollutants (NO₂: +3% and O₃: +5%) but a decreasing trend in FFCO₂ per capita (–14%), while Southeast Asia, East Asia, and Oceania showed the opposite pattern of decreasing air pollution (PM_{2.5}: –20% and O₃: –6%) and increasing FFCO₂ per capita (+22%). The

observed trends in Southeast Asia, East Asia, and Oceania were largely driven by urban areas in China, which constituted 63% of the urban areas in the GBD super-region. Additionally, in the top 20 most populous global cities, Osaka/Kyoto was the only city exhibiting significant decreases in all four pollutants (Supplementary Fig. 3). None of these cities met the WHO AQGs for $PM_{2.5}$ and NO_2 , and most cities except for Quezon City/Manila did not meet O_3 WHO AQG in 2019.

For all six pairwise combinations between the four pollutants, over 50% of urban areas had positive correlations (including both significant and nonsignificant correlations) between pollutant trends globally (PM_{2.5}-NO₂: 62%, PM_{2.5}-O₃: 68%, PM_{2.5}-FFCO₂ per capita: 58%, NO₂-O₃: 57%, NO₂-FFCO₂ per capita: 57%, and O₃-FFCO₂ per capita: 54%, Fig. 4). Over 20% of urban areas had a significant positive correlation (PM_{2.5}-NO₂: 21%, PM_{2.5}-O₃: 26%, PM_{2.5}-FFCO₂ per capita: 20%, NO₂-O₃: 20%, NO₂-FFCO₂ per capita: 24%, and O₃-FFCO₂ per capita: 24%). The highest percentages of positive correlations for PM_{2.5}-O₃ among these pairs were consistent with the findings from the cross-sectional comparison in 2019. When cities were divided into relatively larger versus smaller cities based on population size, using a population threshold of 50,000, the large city group (51% of all urban areas) had a greater proportion of significant positive correlations for every pair compared to all cities combined (PM_{2.5}-NO₂: 33%, PM_{2.5}-O₃: 29%, PM_{2.5}-FFCO₂ per capita: 28%, NO₂-O₃: 22%, NO₂-FFCO₂ per capita: 29%, and O₃-FFCO₂ per capita: 26%, Supplementary Fig. 4B). In contrast, the small city group (49% of all urban areas) showed a lower proportion of significant positive correlations (PM_{2.5}-NO₂: 9%, PM_{2.5}-O₃: 24%, PM_{2.5}-FFCO₂ per capita: 12%, NO₂-O₃: 18%, NO₂-FFCO₂ per capita: 19%, and O₃-FFCO₂ per capita: 21%, Supplementary Fig. 4A).

The GBD super-regions exhibited different correlation patterns (Fig. 4), which generally aligned with the patterns of temporal trends in

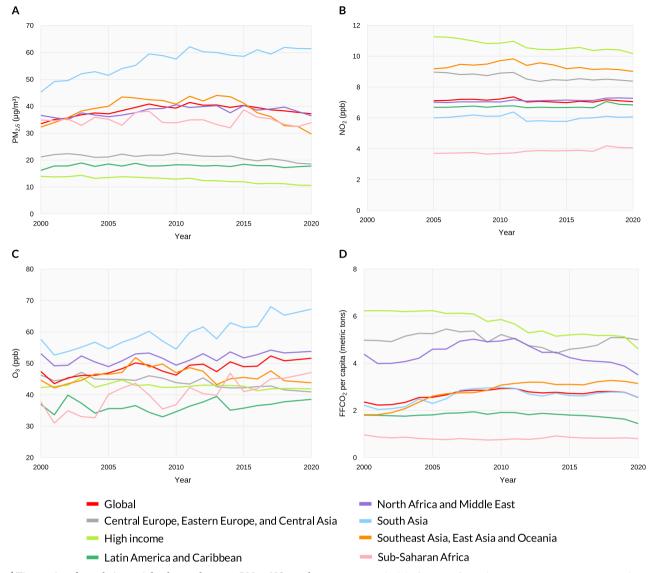


Fig. 2 | Time-series of population-weighted annual average $PM_{2.5}$, NO_2 , and OSDMA8 (O₃) concentrations, and FFCO₂ per capita. A Population-weighted annual average $PM_{2.5}$ (µg/m³, regional mean); n=13,185. B Population-weighted annual average NO_2 (ppb, regional mean); n=12,981. C Population-weighted

OSDMA8 (O₃) (ppb, regional mean); n = 12,941. **D** FFCO₂ per capita (metric tons, regional median); n = 13,182. Only urban areas with full-time series available between 2000 (2005 for NO₂) and 2019 for each pollutant were included.

Fig. 3. In Central Europe, Eastern Europe, and Central Asia and Highincome countries, where overall simultaneous decreases were observed, over 20% of urban areas showed a significant positive correlation for every pair, resulting in high percentages of total positive correlations (including both significant and non-significant positive correlations) ranging from 53% to 92%. South Asia also showed relatively high percentages of urban areas with total positive correlations over 43%. We observed cases that urban areas with negative correlations are more or equally prominent as those with positive correlations, particularly in Latin America and Caribbean and North Africa and Middle East (total positive correlations ranging from 38 to 61%), where coupling trends were rarely found between pollutants in Fig. 3. Southeast Asia, East Asia, and Oceania and Sub-Saharan Africa had mixed patterns depending on pollutants. As Southeast Asia, East Asia, and Oceania showed opposing or decoupling trends between air pollution and FFCO₂ per capita, pairs that included FFCO₂ per capita had higher percentages of total negative correlations than the other pairs (49-61% for pairs with FFCO₂ per capita vs. 20–43% otherwise). In Sub-Saharan Africa, we found higher percentages of total negative correlations for pairs with PM_{2.5} (56-66% for pairs with PM_{2.5} vs. 25-45% otherwise), which was the only pollutant not exhibiting coupling trends with the other pollutants in Fig. 3.

Urban boundary analysis

We next compared urban PM_{2.5}, NO₂, and O₃ concentrations and FFCO₂ per capita using two different urban boundaries: built-up area from the Global Human Settlement Model grid (GHS-SMOD) dataset and administrative boundaries using the C40 Cities dataset (urban boundaries comparison in Supplementary Fig. 5). In 92 urban areas present in both datasets (90 urban areas for O₃ due to missing values in the O₃ dataset), PM_{2.5} and O₃ concentrations showed high agreement between estimates from the two urban boundary definitions, with correlation coefficients (r) near 1.00 and pvalues < 0.0001 (Figs. 5 and 6). NO2 showed lower agreement when compared to PM_{2.5} and O₃, but still high agreement of r = 0.78 (p-value < 0.0001) in 2019 concentration estimates, as well as high agreement in temporal trends for 2005-2019 (r = 0.99 and p-value < 0.0001). We found moderate correlations for FFCO₂ per capita across the two urban boundary definitions in both annual concentration estimates in 2019 (r = 0.67 and p-value < 0.0001) and temporal trends for 2005–2019 (r = 0.49 and p-value < 0.0001). However, the correlation for temporal trends improved when excluding an extreme point having over +300% change from the C40 Cities definition (Fig. 6), resulting in stronger correlation (r = 0.75 and p-value < 0.0001). The C40 Cities definition resulted in higher FFCO₂ per capita than the GHS-

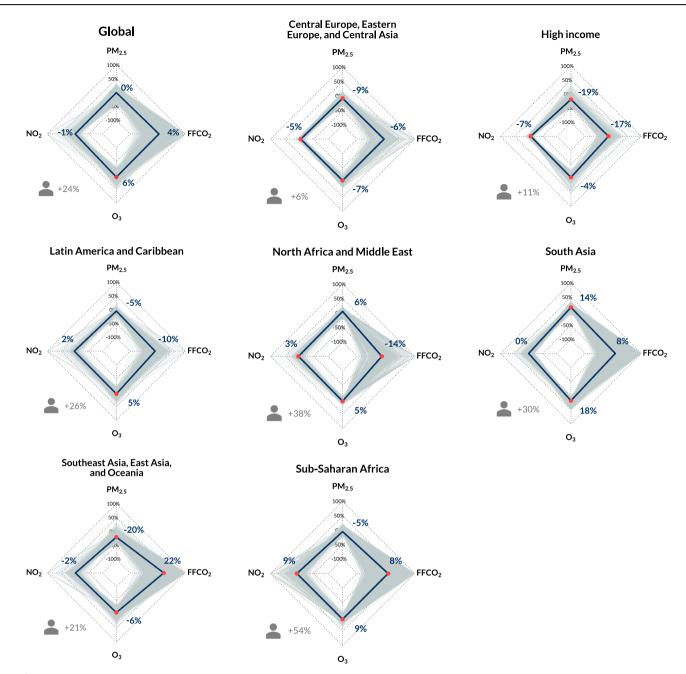


Fig. 3 | Percent changes in population-weighted annual average $PM_{2.5}$, NO_2 , and OSDMA8 (O_3) concentrations and FFCO $_2$ per capita in urban areas from 2005–2007 to 2017–2019. Gray lines indicate percent changes in individual cities located within each region, and navy lines indicate percent changes of averaged values across all cities in each region. Red dots indicate statistically significant monotonic trends between 2005 and 2019. Person icons indicate percent changes in total population from 2005–2007 to 2017–2019. Values above 100% were

consolidated as 100%. Only urban areas with full-time series available between 2005 and 2019 for every pollutant were included. Global: n=12,730; Central Europe, Eastern Europe, and Central Asia: n=605; High income: n=1243; Latin America and Caribbean: n=885; North Africa and Middle East: n=1185; South Asia: n=3858; Southeast Asia, East Asia, and Oceania: n=2789; and Sub-Saharan Africa: n=2165.

SMOD (mean: 3.48 vs 4.49 metric tons and median: 2.19 vs 2.37 metric tons, respectively; Supplementary Table 1).

Differences in estimates of three criteria air pollutants using the two urban definitions did not lead to large differences in the numbers of cities exceeding the WHO interim targets and AQGs (Supplementary Table 2). However, different definitions resulted in different numbers of urban population living in cities exceeding the WHO interim targets and AQGs, even when cities exceeding a guideline were the same. This discrepancy arose because the total population of each city was estimated differently from the two urban boundaries. For example, although all 92 cities exceeded $PM_{2.5}$ AQG, urban population in these cities was estimated as 771 million people

from the GHS-SMOD definition and 548 million people from the C40 Cities definition. Differences in pollutant levels between the two urban definitions in 2019 were correlated with several urban characteristics, including total area, differences in total area, and differences in total population (Supplementary Table 3 and Supplementary Fig. 6). For instance, using an urban definition with a larger total area would likely result in lower urban $\rm NO_2$ concentrations and higher $\rm O_3$ concentrations and FFCO_2 per capita.

Discussion

We developed a globally consistent, multi-pollutant database of population-weighted annual average PM_{2.5}, NO₂, and OSDMA8 (O₃)

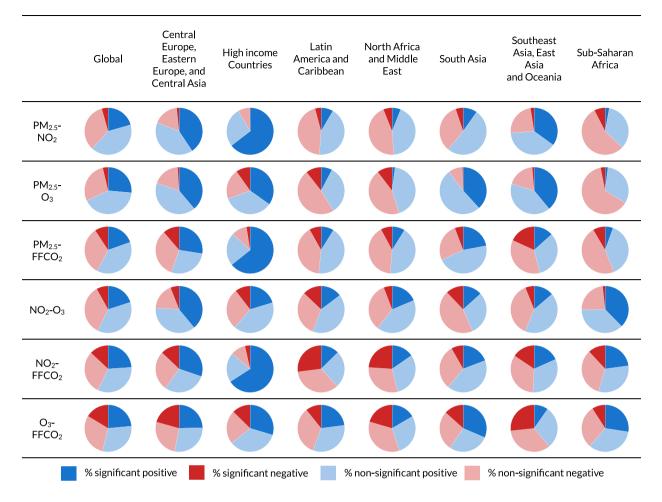


Fig. 4 | Percent of cities with each type of correlation between pollutant trends. Only urban areas with full time series available between 2005 and 2019 for every pollutant were included. Global: n = 12,730; Central Europe, Eastern Europe, and

Central Asia: n = 605; High income: n = 1243; Latin America and Caribbean: n = 885; North Africa and Middle East: n = 1185; South Asia: n = 3858; Southeast Asia, East Asia, and Oceania: n = 2789; and Sub-Saharan Africa: n = 2165.

concentrations and FFCO2 emissions for all urban areas worldwide, which can be utilized for tracking the progress of urban areas toward achieving WHO guidelines and SDGs. Across these urban areas, we found a significant 6% increase in O₃ concentrations globally from 2005-2007 to 2017-2019 and relatively small or no changes in PM_{2.5}, NO_2 , and $FFCO_2$ per capita (+0%, -1%, and +4%, respectively). Across a majority of the global urban areas considered in our study, the four pollutants were positively correlated with one another, reflecting that these pollutants and/or their precursors tend to be co-emitted and have the potential to be reduced simultaneously through mitigation. However, there were substantial variations in temporal trends and correlations by GBD super-region. For example, High-income countries showed significant decreasing trends in all four pollutants and predominantly showed positive correlations across all pollutant pairs. Contrastingly, South Asia and Sub-Saharan Africa showed large significant increases in air pollution and/or FFCO2 per capita during the study period, resulting in high percentages of positive correlations among trends of certain pollutant pairs. In Latin America and Caribbean and North Africa and Middle East, which exhibited decoupling or opposing trends between pollutants, there were similar or even higher numbers of urban areas with negative correlations compared to those with positive correlations.

This study updates urban estimates of air pollution and CO_2 emissions from previous literature by using newer global gridded datasets and reaches broadly consistent conclusions as previous studies. A previous study reported similar $\mathrm{PM}_{2.5}$ trends in urban areas globally, characterized by an initial increase between 2000-2011 followed by a decline²¹. Also, some

studies reported a similar spatial distribution of urban PM_{2.5} concentrations in recent years (2019–2020) across global regions, for example, the highest PM_{2.5} concentrations observed in South Asia^{21,27}. We also observed consistent global and region-specific increases (rapid increases in South Asia and Sub-Saharan Africa) in urban O₃ concentrations with previous studies that used similar datasets and methodologies as this study^{13,18}. For FFCO₂ per capita, although direct comparisons with previous studies were not possible due to variations in data sources and spatial units of estimation (e.g., cities, states, and countries), we found consistent temporal trends with previous studies. Increasing FFCO₂ per capita in Southeast Asia, East Asia, and Oceania and Sub-Saharan Africa as well as decreasing trends in Highincome countries observed in this study were consistently found in previous studies^{28–31}.

However, urban NO_2 trends were inconsistent with previous literature. Globally, we found a 1% non-significant decrease between 2005–2007 and 2017–2019, while Anenberg et al. found a 13% decrease between 2000 and 2019, resulting from a rapid decline after 2010^{20} . This difference might stem from different predictors and statistical methods used by the land-use regression (LUR) models, and ground-level monitoring data used for training the LUR models. We used a recently-developed NO_2 datasets from Larkin et al., which predicted surface NO_2 concentrations at 50 m resolution with Ozone Monitoring Instrument (OMI) column NO_2 and twenty landuse and meteorological variables, trained by 8250 monitors globally³², while Anenberg et al. predicted NO_2 concentrations at 1 km resolution with OMI column NO_2 and ten land-use and meteorological variables, trained by ground-level NO_2 data from 5220 monitors²⁰. Also, when computing

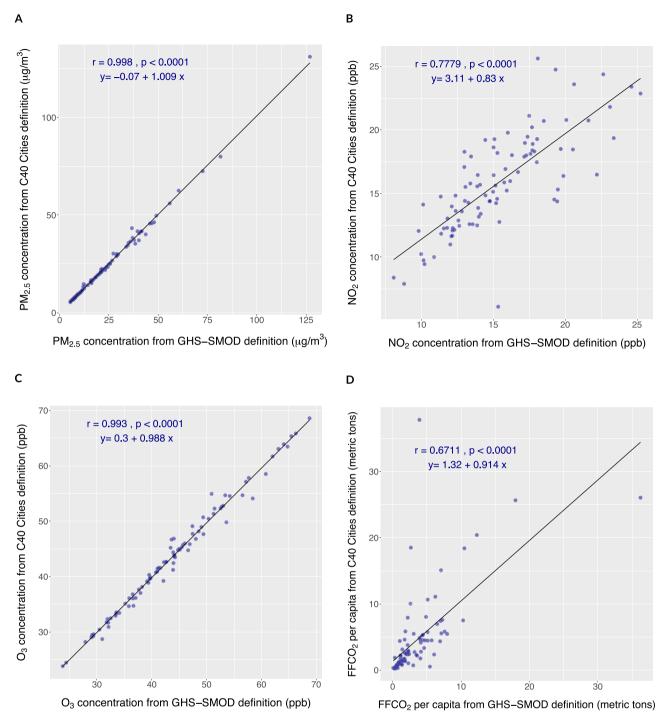


Fig. 5 | Scatter plots of population-weighted annual average PM_{2.5}, NO₂, and OSDMA8 (O₃) concentrations, and FFCO₂ per capita in 2019 derived from two different urban boundary definitions. A Population-weighted annual average

PM_{2.5} (µg/m³); n = 92. **B** Population-weighted annual average NO₂ (ppb); n = 92. **C** Population-weighted OSDMA8 (O₃) (ppb); n = 90. **D** FFCO₂ per capita (metric tons); n = 92.

trends, we calculated the mean of population-weighted annual averages of all urban areas across the globe or a specific GBD super-region (i.e., an average of averages), while Anenberg et al. calculated the population-weighted average of all urban grid cells across each region at once 20 . These differences between studies may collectively lead to different final estimates of urban NO_2 . However, we expect that the new NO_2 estimates from Larkin et al. might provide better estimates as this dataset has a higher prediction performance with an R^2 of 0.63 globally, compared to 0.54 in Anenberg et al. 20,32 , which is likely due to the use of a larger set of predictor variables, finer spatial resolution in the input data, and a greater volume of training data.

We found positive correlations between long-term trends of the four pollutants across the majority of urban areas worldwide. Specifically, we found more conclusive evidence of a correlation between urban $PM_{2.5}$ and O_3 concentrations across different analyses. These findings provide empirical evidence on the co-emission patterns of these four pollutants and/ or their precursors, further stressing the co-benefits of air pollution and climate actions in reducing both air pollutants and CO_2 emissions. Some literature has reported positive correlations between long-term trends of $PM_{2.5}$ and NO_2 concentrations, primarily driven by the contribution of traffic emissions $^{33-35}$. We found stronger evidence of a positive correlation between annual $PM_{2.5}$ and O_3 levels, but the available evidence on the

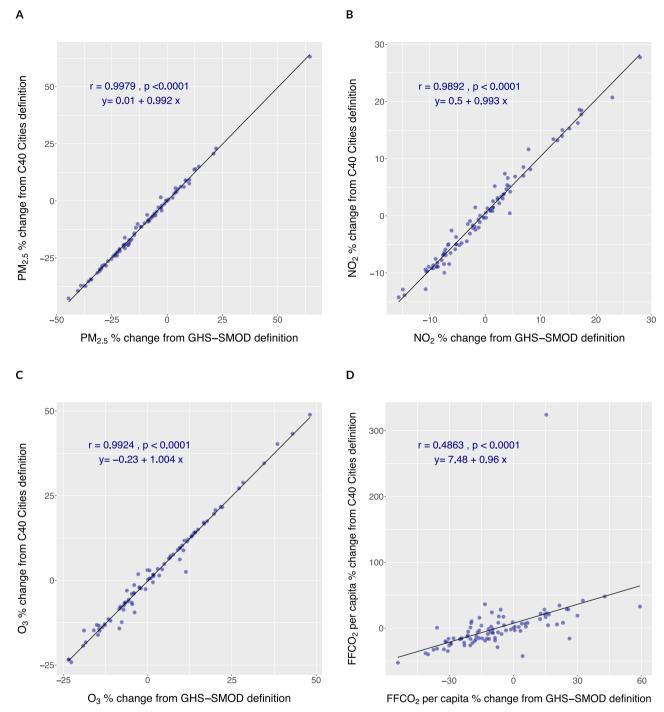


Fig. 6 | Scatter plots of percent changes of population-weighted annual average $PM_{2.5}$, NO_2 , and OSDMA8 (O₃) concentrations, and FFCO₂ per capita from 2005–2007 to 2017–2019, derived from two different urban boundary

definitions. A Population-weighted annual average $PM_{2.5}$ ($\mu g/m^3$); n = 92. B Population-weighted annual average NO_2 (ppb); n = 92. C Population-weighted OSDMA8 (O₃) (ppb); n = 90. D FFCO₂ per capita (metric tons); n = 92.

correlations of long-term averaged O_3 concentrations with the other two air pollutants is mixed and limited $^{35-39}$. Studies reporting positive correlations between O_3 and the other two pollutants suggest that this may be because O_3 precursors, such as nitrogen oxides (NO_x) and volatile organic compounds (VOCs), are closely related to the formation of secondary $PM_{2.5}$ and $NO_2^{35,40}$. Specifically for $PM_{2.5}$, some studies have reported that its correlation with O_3 may arise from their dependency on specific meteorological factors such as temperature inversion and atmospheric stagnation since both pollutants are secondary pollutants (partially for $PM_{2.5})^{38,39}$. However, due to the intricate chemical reactions of O_3 , the evidence remains inconsistent across different spatiotemporal contexts, making it challenging to get

a globally unified interpretation of its correlations with other pollutants. Regarding CO_2 emissions, only a few studies have investigated its relationship with $\mathrm{PM}_{2.5}$ concentration using observational data^{41,42}. Many studies have identified its potential correlations with air pollution only indirectly, inferring from air quality models assuming their co-emissions from common sources^{26,43,44}.

Given the common emission sources among the pollutants and/or their precursors, we hypothesized that we might uncover consistently positive correlations among these pollutant trends. Although we found that positive correlations dominate in many cases, positive correlations were not always dominant, particularly in urban areas in Latin America and Caribbean and North Africa and Middle East (Fig. 4). Long-term trends of air pollution are not exclusively influenced by local emissions, but also by various factors like meteorological conditions, natural emissions, and longrange transport, with variations arising from the distinct formation characteristics of different pollutants. For example, reducing NOx emissions may decrease NO2 and PM2.5 concentrations but could leave O3 levels unchanged or even increased due to complexities in O3-NOx-VOC chemistry⁴⁵. Climate change and rising temperature also affect air pollutant emissions from natural sources, such as increased wildfire activities⁴⁶, which release air pollutants independently of anthropogenic emissions, unlike FFCO₂. The impacts of climate change can vary across air pollutants; for instance, natural sources of O₃ precursors include biogenic VOCs, soil NOx, wetland methane, and wildfires⁴⁷, whereas PM_{2.5} is more heavily impacted by wildfires and natural dust⁴⁸. Additionally, the fact that O₃ formation accelerates at higher temperatures⁴⁹, along with increased emissions of its precursors from natural sources, could explain why O₃ concentrations showed significant increases globally across all four pollutants. Lastly, pollutants like PM2.5 and O3 can be transported across regions and even hemispheres by large-scale air circulation, as O₃ has a relatively long atmospheric lifetime and both pollutants have long-lived precursors^{47,50,51}. As a result, long-range transport can either offset or amplify the impacts of local emission changes in certain areas.

While the pollutants considered in our study and/or their precursors are co-emitted, long-term trends of air pollutants and CO₂ emissions can be independently influenced by different emission reduction strategies or exogenous events (e.g., armed conflicts and migration)^{22,52,53}. For example, while renewable and nuclear energy can effectively mitigate both air pollutants and FFCO₂ emissions, carbon capture and storage have no impacts on improving air quality but effectively mitigate FFCO₂ emissions, and motor vehicle tailpipe controls can only mitigate air pollution²². Therefore, further studies are needed to explain how different combinations of mitigation strategies and regional events influence long-term trends of each pollutant, particularly in areas showing opposing and decoupling trends among pollutants like Latin America and Caribbean and in North Africa and Middle East.

The variations in trends and correlation patterns we found across GBD super-regions likely stem from differences in primary sources of pollutants and regional environmental and socioeconomic contexts. For instance, urban areas in High-income countries showed significant decreases in all four pollutants and positive correlations between pollutant trends. These findings may reflect the impacts of comprehensive air pollution control strategies, active support mechanisms for reducing GHGs emissions, and their synergistic impacts in High-income countries, including many Western European, North American, and East Asian countries. For example, the US has regulated criteria air pollutants under the Clean Air Act and has strengthened the regulations continuously over time, incorporating various strategies like establishing emission standards for primary emission sources. Additionally, federal- and state-level GHGs mitigation actions, including renewable portfolio standards, increased investment in research and development, and renewable energy tax credits, have supported the transition to renewable energy in the energy sector^{54–56}. This transition, coupled with a simultaneous increase in natural gas use, replacing coal⁵⁷, has resulted in reductions of both total emissions and per capita emissions of FFCO₂⁵⁸. Also, as several measures overlap between air pollution and climate actions, such as increasing the use of renewable energy and cleaner fuels and increasing motor vehicle fuel efficiency, these overlapping measures might contribute to complementary reductions of both air pollution and CO2 emissions^{22,59}.

On the other hand, urban areas in South Asia and Sub-Saharan Africa experienced large increases in air pollution and/or FFCO₂ per capita, resulting in largely positive correlations in their temporal trends. Such trends might be attributable to rapid economic growth in these regions as these two regions experienced among the largest economic and population growths, leading to large increases in fossil fuel use in transportation, industrial sources, and power plants sectors⁶⁰⁻⁶². While South Asian

countries have begun controlling emissions of air pollutants and GHGs during the study period by setting emission standards and transitioning to cleaner energy, especially focusing on vehicle emissions, the influence of these actions was likely outweighed by rapid urbanization and economic growth^{63,64}. Some studies have suggested that the lack of public awareness, appropriate air quality monitoring, political support, and international cooperation could also lead to the ineffectiveness of the control measures in these regions^{63,65}. In Sub-Saharan Africa, as urbanization is still in its early stages and household air pollution has been a major focus in policies, there has been less action addressing ambient air pollution, likely combined with a lack of political will and capacity 62,66. As a result, the region might have been unable to effectively counter the increasing emissions from economic growth and urbanization. Some studies have suggested various policy options for countries in these regions, which decouple air pollution and GHGs emissions from economic growth while not impeding growth itself, including improved energy efficiency, public transport, waste management, and increased use of cleaner cook stoves^{64,67}. By adopting these strategies, these regions might effectively achieve both developmental and environmental goals simultaneously.

We compared two urban boundary definitions-from GHS-SMOD and C40 Cities-both of which have important roles in research and policy. The GHS-SMOD is one of the most frequently used urban boundary datasets in scientific research that provides a globally consistent characterization of urban areas. On the other hand, the urban boundary datasets from the C40 Cities may better reflect the scale at which city-level policies are implemented. We found that the different urban definitions influenced urban NO₂ concentrations and FFCO₂ per capita in 2019, while PM_{2.5} and O₃ concentrations were largely insensitive to urban definitions. However, the agreement was still at moderate to high levels with correlation coefficients over 0.6. Several studies have reported variations in urban estimates of air pollution and CO₂ emissions by different urban definitions⁶⁸⁻⁷¹. Discrepancies in NO₂ estimates from different urban definitions likely reflect its high intraurban spatial heterogeneity due to localized pollution hotspots and short lifetime^{72,73}. Similarly, FFCO₂ per capita can vary with urban definitions, as emission estimates are directly derived from the geographic distribution of emission sources, which may differ between city centers and suburbs-for instance, suburbs often have more large point sources than city centers^{71,74}. Also, certain sources may emit different amounts of CO₂ emissions between urban centers and suburbs, which can make urban CO₂ emission estimates sensitive to urban boundary definitions. For example, several studies found that people living in suburbs are more likely to have higher commuting CO₂ emissions than city centers due to longer-distance commuting travel^{74,75}. Therefore, in future studies, particularly those focused on NO₂ concentrations and CO₂ emissions, researchers should use urban definitions that are appropriate for their research objectives and exercise caution when comparing findings with other studies that might have relied on other urban definitions.

This study has some limitations. First, while this study utilized recentdeveloped global datasets of air pollution and GHGs incorporating state-ofthe-art methods to improve the accuracy of measurements, these datasets may still contain errors due to insufficient input data, incorrect model processes, and erroneous emission data^{20,76–78}. Second, the level of prediction errors in the air pollutant datasets can differ by global regions. Global estimates of PM_{2.5}, NO₂, and O₃ concentrations incorporate to varying degrees ground-level monitors that are predominantly located in Highincome countries within the Global North, and estimated concentrations are likely more uncertain for unmonitored urban areas in the Global South. Van Donkelaar et al. and DeLang et al. reported the largest uncertainty estimates of global PM_{2.5} and O₃ concentrations, respectively, in undermonitored regions, as shown in Supplementary Table 476,77. Accordingly, we observed the lowest absolute uncertainties in High-income countries when compared to the other regions for both PM_{2.5} and O₃ (Supplementary Table 4). We expect that using satellite data with complete global coverage used as inputs in these datasets might ameliorate this potential for PM_{2.5} and NO₂. However, direct satellite observations are unavailable for O₃ due to the

high abundance of O_3 in the upper atmosphere, making O_3 estimates in regions without monitoring stations and O_3 distributions at the fine spatial scale rely primarily on bias-corrected models rather than direct observations. Third, additional uncertainty may also arise from the population dataset due to the absence of consistently available sub-national census datasets at fine scales and the inaccuracy in those source datasets^{79,80}.

Fourth, our results may be influenced by the choice of spatial resolution in the prediction models. In our study, O₃ and CO₂ datasets have a coarser resolution of 10 km, compared to PM_{2.5} (1 km) and NO₂ (50 m). Several studies found that coarse model resolutions that fail to capture smaller-scale variations in air pollutant concentrations can underestimate pollution levels due to the smoothing of hotspots⁸¹⁻⁸⁴. Also, many urban areas are represented by only a single or a few grid cells for O₃ concentration and FFCO₂ per capita (92% of urban areas have total areas smaller than 100 km²). While some studies found no significant improvement in accuracy with resolutions finer than 10 km for O₃, given its nature as a secondary pollutant with less spatial variation^{85,86}, CO₂ emissions can vary substantially at small scales due to its dependence on specific locations of emission sources. For PM_{2.5} and NO₂, although, to the best of our knowledge, the datasets we used have the finest resolutions among existing global models, it still remains uncertain whether these resolutions are fine enough to capture small-scale variations. Moreover, fine model resolutions may sometimes compromise predictive power and require additional computational costs⁸⁵. For example, the NO₂ dataset has lower prediction power than the other two pollutants, with R2 of 0.84, 0.63, and 0.81 for PM_{2.5}, NO₂, and O₃, respectively. Therefore, while finer model resolutions are essential to resolve fine-scale urban gradients of the pollutants, researchers should carefully weigh the trade-offs between achieving finer resolutions and the associated computational demands and potential impacts on model performance. Lastly, the GHS-SMOD urban boundary definition we used is from 2015 estimates⁸⁷. As urban areas have been expanding rapidly, especially those in the Global South experiencing rapid urbanization, their boundaries might have changed prior to or following 2015. However, to our knowledge, the GHS-SMOD is the only dataset to provide global coverage of all urban areas using consistent input datasets and methodologies. Additionally, using temporally varying urban boundary definitions would make it difficult to distinguish the impacts of urban actions that reduce emissions and pollution from those of city expansion or diminution. Therefore, we used 2015 GHS-SMOD urban boundary definitions.

Despite these limitations, we provide one of the first systematic comparisons of global trends of PM_{2.5}, NO₂, and O₃ concentrations and FFCO₂ per capita in urban areas. Given that urban areas have been hubs of air pollutants and GHG emissions, as well as the focus of diverse mitigation actions undertaken globally, our findings highlight the outcomes of past urban initiatives and provide insights for future strategies in different global regions. By examining potential correlations between pollutant trends globally and across various regions using datasets that are based on historical observations, we provide empirical evidence on ways that air pollution and GHGs emissions are coupled and might be simultaneously mitigated. The regional variations in the results, such as the contrasting trends between High-income countries and South Asia or Sub-Saharan Africa, stress the need to further understand the local and regional drivers of air pollution and GHGs emissions and tailored region-specific strategies. Furthermore, the influence of different urban boundary definitions on pollutant estimates underscores the importance of how we define urban air pollution and CO₂ emissions in research and policy.

Methods

Population-weighted annual average PM_{2.5}, NO₂, and OSDMA8 (O₃) concentrations and FFCO₂ per capita

We leveraged existing global, highly spatially-resolved datasets of $PM_{2.5}$, NO_2 , and O_3 that represent the current state-of-the-science estimates of each pollutant. Specifically, we used a global $PM_{2.5}$ dataset for 2000–2020 from van Donkelaar et al.⁷⁷, which estimates annual average concentrations at 0.01° (~1 km) resolution, combining satellite remote sensing of aerosol

optical depth, a chemical transport model, and ground-based measurements. For NO₂, we employed a global dataset for 2005–2020 from Larkin et al. This dataset provides annual average concentrations at 50 m resolution using a land-use regression model that ingests remotely-sensed measurements from the OMI with other land use variables, trained against ground-based measurements worldwide³². For O₃, we used a global dataset for 2000–2020 from the GBD 2019 study⁸⁸, which is extrapolated from an O₃ dataset of Delang et al.⁷⁶. This dataset estimated the annual maximum of the six-month running mean of the monthly average daily maximum 8 h mixing ratio (ozone season daily maximum 8 h mixing ratios, OSDMA8) at 0.1° (~10 km) resolution combining ground-based O₃ observations from the Tropospheric Ozone Assessment Report (TOAR) and the Chinese National Environmental Monitoring Center Network, and outputs from multiple global atmospheric models^{76,88}.

Evaluations of these datasets against ground-based observations revealed a high degree of agreement (i.e., R^2 of 0.84, 0.63, and 0.81 for $PM_{2.5}$, NO_2 , and O_3 , respectively) and improved predictive performance over previous global estimates of these pollutants 32,76,77 . Also, by using the annual averaged $PM_{2.5}$ and NO_2 concentrations and the annual maximum of the six-month running mean for O_3 , which is likely to include mostly warm season concentrations, we effectively controlled day-to-day and seasonal variations of air pollution, and thus these estimates are likely to reflect the impacts of long-term emission changes.

To calculate FFCO₂ per capita, we used annual sector-specific emission datasets of the Emission Database for Global Atmospheric Research (EDGAR) v8.0 for 2000-2020, which has 0.1° (~10 km) resolution with global coverage89. The EDGAR dataset contains emissions of multiple GHG species, but we only considered CO₂ emissions emitted from anthropogenic sources within each grid cell. This dataset provides production-based CO2 emissions, which account for CO₂ emissions in the location where goods and services are produced, rather than where they are consumed. For example, even if a certain amount of CO₂ is emitted from producing goods that are exported and used outside a grid cell, the emissions are still accounted for in the total CO2 emission of that grid cell. We defined FFCO2 emissions as a sum of total emissions from all sectors of anthropogenic sources, including power industry, industrial combustion and processes, buildings, transport, agriculture, fuel exploitation, and waste¹⁹. While PM_{2.5}, O₃, and FFCO₂ per capita are available annually from 2000 to 2020, NO₂ estimates are only available annually from 2005 to 2020 because OMI, which provides remotely-sensed NO₂ measurements used by the land-use regression model used to estimate surface-level NO₂, was launched in 2004.

To compute the urban estimates, we used 2015 GHS-SMOD urban boundaries for 13,189 urban areas worldwide⁸⁷. The GHS-SMOD urban boundaries were defined by the European Commission based on global geographic distribution of populations and urban structures⁸⁷. We additionally obtained global gridded population counts for 2000–2020 at 1 km resolution from WorldPop, which incorporates country-specific population estimates and population-related geospatial factors⁹⁰. We regridded the air pollutant and CO_2 datasets to the same 1 km resolution and extent as WorldPop. For each urban area defined by the GHS-SMOD, we calculated population-weighted average of $PM_{2.5}$ (gridded annual average), NO_2 (gridded annual average), and O_3 (gridded OSDMA8) concentrations, as well as FFCO₂ per capita. Population-weighted average concentrations were calculated using Eq. (1):

$$c_{i,population-weighted} = \sum_{j} (p_{i,j} \times c_{i,j}) / \sum_{j} p_{i,j}$$
 (1)

where c represents pollutant concentration, i denotes an urban area, j denotes a grid cell in urban area i, and p represents population count. FFCO₂ per capita was calculated by dividing the total FFCO₂ emissions in an urban area by the total population of that urban area. In the calculations, grid cells with missing values in either pollutants or population datasets were excluded. Missing grid cells were primarily located over oceans and inland water bodies. The total number of cities varies by year and pollutant due to

differences in the pollutant datasets, and details on these variations are provided in the figure captions.

Temporal trends for 2005-2019

Urban population-weighted annual average PM2.5, NO2, and OSDMA8 (O₃) concentrations and FFCO₂ per capita were aggregated globally and across GBD super-regions for every year by calculating mean values of air pollutant concentrations and median values of FFCO2 per capita overall urban areas within each region. For FFCO₂ per capita, because of the high variation and extreme values, we used the median as a summary statistic instead of the mean. The GBD super-region categorization groups countries into seven large regions based on geographic proximity and similarity of epidemiological patterns (Supplementary Fig. 1)⁹¹. While we visualize trends in pollutants from 2000 to 2020 (2005–2020 for NO₂) (e.g., Fig. 2), our statistical analysis of trends (e.g., Fig. 3) is based on 2005-2019 due to NO₂ data availability starting from 2005 and 2019 being the latest year unaffected by the COVID-19 pandemic. In the main analyses, we summarized temporal trends as percent changes in pollutant levels by comparing the 3-year average of annual means/medians from 2005 to 2007 to that from 2017-2019. Here, we used 3-year averages to minimize the influence of outliers from any single year. Additionally, the significance of monotonic temporal trends between 2005 and 2019 was tested using the Mann-Kendall test.

Then, we calculated the Pearson correlation coefficient (r) characterizing the temporal correlations among population-weighted annual average PM_{2.5}, NO₂, and OSDMA8 (O₃) concentrations and FFCO₂ per capita. These coefficients were calculated at the city level, which resulted in six different correlation coefficients of every possible pair of the four pollutants for every individual city, and then were summarized at GBD super-regions and globally. At the aggregated level, we present the proportions of urban areas with four categories of correlation coefficients: significant positive, non-significant positive, significant negative, and non-significant negative. Statistical significance for all statistical tests in this study was set at the 95% confidence level, where a p-value less than 0.05 was considered statistically significant.

Urban boundary analysis

We tested the influence of two different urban boundary definitions on our estimates of the four pollutants: the GHS-SMOD definition, which was used for our primary results as this is the most frequently used definition in global-scale research^{13,20,21,92,93}, and the C40 Cities definition. As stated before, the GHS-SMOD urban boundaries were defined based on global geographic distribution of populations and urban structures⁸⁷. The C40 Cities definition relies on self-reported urban boundaries from their 97 member cities, and boundaries for a particular city might include broader geographic areas (e.g., suburbs, exurbs) than just the immediate urban areas. We compared annual level estimates of the four pollutants in 2019 and temporal trends for 2005-2019 from the two different definitions, across 92 urban areas (90 urban areas for O₃ due to missing values in the O₃ dataset) represented by both GHS-SMOD and C40 Cities. For temporal trends, we used the same percent change metric as in our temporal trends analysis. Additionally, to understand how differing urban boundary definitions can influence public health implications regarding urban air pollution, we compared the numbers of cities exceeding the WHO AQGs and the population living in those cities, estimated from the two urban definitions.

To identify potential drivers of the differences in pollutant estimates between urban boundary definitions, we correlated the differences in pollutant estimates between GHS-SMOD and C40 Cities definitions with several urban characteristics. These characteristics include total area, total population, population density from the GHS-SMOD, the difference in population and area between GHS-SMOD and C40 Cities, and Moran's I. Here, Moran's *I* characterizes the level of spatial autocorrelation of pollutant estimates within a given city included in the GHS-SMOD. We correlated the differences in pollutant levels with these metrics using Pearson and

Spearman correlations depending on the distribution of the variable (Supplementary Table 3).

Data availability

The datasets generated during the current study are available in CSV format from the author's GitHub repository (https://github.com/sooyeon-kim-29/ Tracking-air-pollution-and-CO2-emissions....git) and GW Air, Climate, and Health lab's website (https://blogs.gwu.edu/sanenberg/pm2-5-no2-and-ozone-data-for-13000-cities-worldwide/) with online data visualization tools. $PM_{2.5}$ datasets used in the current study are available via the Washington University Atmospheric Composition Analysis Group website (https://sites.wustl.edu/acag/datasets/surface-pm2-5/), and O_3 datasets are available through our co-author's repository on Zenodo (https://zenodo.org/records/8320001).

Code availability

The code used for the current study is available from the author's GitHub repository (https://github.com/sooyeon-kim-29/Tracking-air-pollution-and-CO2-emissions...git).

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Competing interests

The authors declare no competing interests.

Additional information

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