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Key Points:

- Column CO (X_{CO}) trends vary regionally by 4%–5% year⁻¹ on 5-year scales with an overall global decrease
- Large CO emitting events can cause X_{CO} anomalies of +20 ppb (~20%) compared to other years over thousands of kilometer
- Urban trends are similar enough to regional trends that it is difficult to distinguish them with this system

Supporting Information:

- Supporting Information S1

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Regional and Urban Column CO Trends and Anomalies as Observed by MOPITT Over 16 Years

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Abstract Atmospheric carbon monoxide (CO) concentrations have decreased since the beginning of the century, and the rate of decrease depends on the region. Depending on how regions are chosen, their boundaries may not always align with where there are differences in trends. To address this, we calculate trends within $0.4^\circ \times 0.4^\circ$ grid cells independently throughout the globe using satellite CO retrievals from the Measurements Of Pollution In The Troposphere (MOPITT) satellite instrument from 2002 to 2017. These trends are found with the caveat that there are large singular biomass burning events somewhere nearly every year, and we include examples of large column CO anomalies during sporadic but large burning events in the North American and Eurasian boreal forests, the Amazon, Africa, and Indonesia. CO trends behave similarly within regions that span about a few thousand kilometers. Using TransCom region definitions, we find average trends between -0.9 and 0.1 ppb year⁻¹ (about -0.9 to 0.1% year⁻¹) for 2002–2017. Over 5-year subsets, trends in TransCom regions vary between -3.6 and 1.8 ppb year⁻¹. This substantial spatial and temporal variability in trends is in agreement with other studies. With an understanding of regional trends, we compare with trends from urban areas. Generally, CO trends within urban areas are indistinguishable from regional trends. This may be because of a combination of noise in the data, the large footprint for MOPITT, or because anthropogenic CO reduction measures were implemented before the MOPITT record began. We provide case studies for a few cities, such as Los Angeles, and find long-term variation in the rate of change of column CO.

Plain Language Summary Carbon monoxide (CO) is an atmospheric pollutant produced from combustion, such as wildfires and gasoline powered automobiles. We use a 16-year satellite record to determine how atmospheric CO levels have been changing this century. Though sporadic, large wildfires happen somewhere every year which can affect trends in CO. We find trends for small regions ($\sim 40 \times 40$ km) over the full 16 years and over 5-year subsets. Trends vary significantly among regions and among the 5-year subsets. We then examine how CO levels in cities change compared to their surrounding region. In most cases, CO trends in cities do not vary significantly from trends in the surrounding region. In some regions where we would expect a decrease (like Los Angeles and Mexico City), we find large intra-annual variability.

1. Introduction

Carbon monoxide (CO) is a tropospheric pollutant that is a useful tracer of atmospheric transport and an indicator of emission sources. The combined global budget from direct sources and in situ oxidation is about 2,600 TgCO year⁻¹ (Zheng et al., 2019). Of this, slightly less than half ($\sim 45\%$) is from primary emissions from incomplete or inefficient combustion, split at about 60% fossil fuel and 40% biomass burning. The other half comes as secondary emissions from the oxidation of volatile organic compounds (VOCs) in Earth's atmosphere. Methane (emitted from a variety of sources including natural gas activities, and methanogens under hypoxic conditions) accounts for about two-thirds of secondary CO emissions, with isoprene (primarily emitted from vegetation) also being a major contributor (Zheng et al., 2019). Changes in CO

Table 1
Approximate CO Emission Factors by Source

Source	gCO kg ⁻¹ fuel	References
NZ manufacturing diesel	3	Metcalf and Sridhar (2018)
NZ manufacturing coal	20	Metcalf and Sridhar (2018)
NZ natural gas electricity	1.6	Metcalf and Sridhar (2018)
NZ natural gas residential	1.1	Metcalf and Sridhar (2018)
NZ wood heating	140	Metcalf and Sridhar (2018)
NZ aircraft	10	Metcalf and Sridhar (2018)
NZ diesel vehicles	0.1	Metcalf and Sridhar (2018)
US gasoline vehicles 1980 ^a	250	US EPA (1985)
1990 ^a	150	US EPA (1985)
2000	50	Bishop and Stedman (2008)
2010	20	Bishop and Stedman (2008)
US motorcycles ^b	350	US EPA (1985)
US lawn mowers	600	US EPA (1985)
Wildfires ^c	61–135	Urbanski (2014)

Note. Values are often approximated from references. Though we report the source country (NZ or US) emission factors are expected to be similar for other countries with similar technology.

^aUsing 17 MPG and 41 gCO mi⁻¹ for 1980 and 25 MPG and 17 gCO mi⁻¹ for 1990. ^bSee Table 1.8.2A in reference. Assuming 50 MPG. ^cVaries by up to about a factor of 2 depending on combustion type (e.g., flaming vs. smoldering) (Urbanski, 2014).

may be reflective of changes in combustion efficiency, usage of catalytic converters, number of sources, and emissions of VOCs. At high enough concentrations, CO is a health concern, and it is also a precursor to ozone which can cause health issues at lower concentrations. In addition, CO can be used as a proxy for urban health. Since it is an indicator of inefficient incomplete combustion, it may be coemitted with VOCs which may react and form secondary particulates or may be coemitted with primary particulates.

CO is known to be decreasing globally since at least the beginning of this century (Worden et al., 2013). The decrease has been larger in the Northern Hemisphere (NH) than the Southern Hemisphere (SH), though concentrations are also higher in the NH. Worden et al. (2013) showed a particularly strong decreasing trend over eastern China. Yin et al. (2015) found a significant decrease in CO emissions from China, which was particularly interesting since bottom-up inventories suggested increasing CO emissions. They suggested this was due to technology improvements, despite an increase in amount of fuel burned. Because the column CO over China increased from 2001 to 2004, the decrease from 2004 to 2015 was more significant. Decreasing CO in the NH has been due to decreasing primary sources, rather than decreasing secondary sources or an increasing sink (primarily the OH radical) (Jiang et al., 2017). Compared to 2001, there have been decreases in CO over the United States, Europe, and China. Jiang et al. (2017) also found increasing emissions from India and southeast Asia. Jiang et al. (2018) examined CO and nitrogen oxides (NO_x) trends over the United States and found a slowdown of emission reductions which they attributed to diminishing returns from improvements to gasoline vehicles which have already had significant reductions in CO emissions (see e.g., Table 1).

Measurements of Pollution In The Troposphere (MOPITT) space-based data have been used to understand column CO behavior in a number of regions. Recently, P. Wang et al. (2018) examined differences in regional and urban trends in MOPITT column CO, especially over Eurasia. They found a significant decreasing trend in Moscow from 1998 to 2014 ($3.73\% \pm 0.39\% \text{ year}^{-1}$). Part of the decrease was attributed to a more widespread decrease across Eurasia from 1998 to 2014, with nearby sites decreasing by 0.9–1.7% year⁻¹. They attributed the overall decreasing trend near Moscow to transitioning from domestic to imported cars with catalytic converters. Recently, column CO trends were again shown to have decreased over China during 2003–2017, with no significant trends over India and Indonesia (Zhang et al., 2020).

There have been several studies that have focused on urban CO behavior using MOPITT observations. Worden et al. (2012) used MOPITT CO with a WRF-Chem model to quantify the impact of emission controls for the 2008 Beijing Olympics. Another method is to rotate column CO measurements upwind and downwind of an urban center to help account for urban versus regional behavior (Pommier et al., 2013). This method involves oversampling the observations to obtain smooth spatial distributions of column CO. Oversampling by averaging all soundings within an area will reduce the enhancement of a pollutant in the downwind compared with upwind but should have less of an effect on relative changes with time. Dekker et al. (2017) applied this same method to estimate relative trends and also used a $10 \times 10 \text{ km}^2$ atmospheric model to estimate fluxes from Madrid. They developed error budgets for both methods and examined the impact of a priori emissions, averaging kernel (AK) trends, filtering, and resolution. They concluded that previous uncertainty estimates were too optimistic.

While trends in regional CO and urban CO have been studied before, an objective of this study is to understand the spatial extent of regions that have similar long-term CO changes. Previous trend analysis has often relied on studying regions with boundaries that are well chosen geographically (e.g., Jiang et al., 2017) but may not always distinguish between regions with different long-term trends. We are interested in not

only spatial heterogeneity but also temporal heterogeneity (e.g., Jiang et al., 2018). A second objective is to understand trends on urban scales for a large number (500) of cities, and this study considers urban trends in the context of regional trends.

This study relies on MOPITT observations from 2002 to 2017, described along with ancillary ground-based observations around Los Angeles, and urban databases in Section 2. Our methods are described in Section 3. In Section 4, we analyze gridded anomalies for single events and trends across the globe on various time scales for different regions. Our urban analysis is described in Section 5 and includes a focus on select cities. We conclude and summarize our findings in Section 6.

2. Data Sources

2.1. MOPITT

The MOPITT instrument aboard the Terra satellite launched in December 1999 measures column CO globally. Terra is in a sun-synchronous orbit with a local daytime descending crossing time of around 10:30 (data from the nighttime ascending crossing at 22:30 are not used in this study). MOPITT is still operational and has made the longest satellite record of atmospheric CO (Deeter et al., 2017). With a 600-km swath width, daytime measurements from MOPITT cover nearly the entire globe every 4–5 days. Individual soundings are $\sim 22 \times 22 \text{ km}^2$, and retrievals are performed only for cloud-free scenes. Because of an instrument configuration change in 2001, we use observations from 2002 to 2017 (Drummond et al., 2010). We use the daytime version 7 joint (thermal infrared (IR) and near-IR) L2 column-averaged product with the filters and bias corrections recommended by Hedelius et al. (2019). Because of low near-IR signal over water, the retrieval algorithm only uses the thermal IR band for these soundings. This algorithm is used to obtain a best estimate of an atmospheric CO profile. From this profile, column-averaged dry-air mole fractions of CO (X_{CO}) are calculated.

2.2. Ground-Based Observations

Part of our analysis relies on total column observations of CO from ground-based solar-viewing spectrometers. The Jet Propulsion Laboratory (JPL) MkIV spectrometer has been used to make CO measurements both within the Los Angeles (LA) basin and at other locations going back to 1985 (Toon, 1991). Measurements are made about once or twice a week for about 2 h per day. In addition, measurements have been made from a number of Total Carbon Column Observing Network (TCCON) instruments at two locations inside the basin (Pasadena, ci, 240 m and JPL, jc/jf, 390 m), and one in the less polluted Mojave desert (Armstrong, df, 700 m) (Hedelius et al., 2017; Iraci et al., 2016; Wennberg et al., 2016; Wennberg, Wunch, Roehl, et al., 2014; Wennberg, Wunch, Yavin, et al., 2014; Wunch et al., 2011). These measurements go back as far as July 2007. Usually, measurements with these TCCON instruments are made from sunrise until sunset on all days the system is not under maintenance, weather-permitting.

The GGG software package is used to retrieve X_{CO} from clear-sky observations by both types of instruments (Wunch et al., 2015). For MkIV measurements, we subtract the column H_2O from the whole air column based on surface pressure to calculate a dry-air column (equation 2 of Wunch et al. [2016]).

2.3. Urban Database

The 500 most highly populated urban locations are from the Global Human Settlement Urban Centre Database (GHS-UCDB, Florczyk et al., 2019). This database contains locations for over 13,000 urban areas along with selected attributes. GHS-UCDB defines urban areas based on connected 1 km^2 grid cells each with a population of at least 1,500 or at least 50% built-up surface area totaling 50,000 inhabitants or more. We focus our analysis on the 500 urban areas with the highest populations. Together, these areas include 1.66 billion people and have a population range of 0.95–40.6 million each. These urban areas can each contain several agglomerated municipalities or cities. However, we sometimes refer to an agglomeration as a singular city.

Population density maps are based on the Gridded Population of the World, Version 4 (GPWv4) revision 11 data set for 2015 (Center for International Earth Science Information Network - CIESIN - Columbia University, 2018) (last access: May 31, 2019).

2.4. Emissions Inventory

We obtain emission estimates of CO from 2002 through 2012 from the Emissions Database for Global Atmospheric Research (EDGAR) version 4.3.2 (<http://edgar.jrc.ec.europa.eu/overview.php?v=432>, https://data.europa.eu/doi/10.2904/JRC_DATASET_EDGAR). This is a $0.1^\circ \times 0.1^\circ$ spatial resolution inventory with annual granularity. EDGAR does not include emissions from large scale biomass burning or CO production from VOCs (Crippa et al., 2018). EDGAR is based on numerous assumptions and other underlying data sets whose uncertainties could propagate into the inventory.

3. Methods

3.1. Gridded Global MOPITT Products

We create a global gridded ($0.4^\circ \times 0.4^\circ$) or $\sim 40 \times 40$ km product of monthly averaged data from MOPITT observations, using the squared inverse of the reported errors as weights. This choice of grid was based on the work of Worden et al. (2012) who looked at the change in CO over Beijing before and during the 2008 Olympics. For plotting global maps, we coarsen the grid to $\sim 1^\circ \times 1^\circ$ to reduce figure file size.

From this gridded product, we calculate average values for each month across the 2002–2017 time period by taking the average of a particular month across all years. From these average values, we calculate anomalies by subtracting the mean from the monthly product of a specific year.

We examine trends in the $0.4^\circ \times 0.4^\circ$ monthly averaged product by independently finding the trend in each cell with the Theil-Sen slope estimator (Hollander et al., 2014). Theil-Sen is used instead of least squares to reduce the impact of outliers due to single events or measurement uncertainty. Confidence intervals (CIs, 95%) are determined using a bootstrap method (Efron & Gong, 1983) over monthly averages with 1,000 repeats. However, we assume a Gaussian distribution and convert these to standard deviation. Generally, we assume that a trend does not significantly differ from zero if it is within one standard deviation. Trends are only reported if at least 50 months (of 192) are used. Along with trends, overall average values are also shown in Figure 3. Trends are also found for 5-year windows (for $n \geq 15$ months) and 5-year windows for separate seasons ($n \geq 8$). Specifically, we analyze 2002–2006, 2005–2009, 2009–2013, and 2013–2017.

Similar grids of the means and trends are created from the EDGAR inventory. Because the version we use only goes through 2012, we analyze temporal subsets using years 2002–2006, 2005–2009, and 2009–2012 along with the overall 2002–2012 trend.

3.2. Urban Versus Regional Differences

We examine how X_{CO} trends measured using MOPITT observations may differ in an urban area compared to a larger region, with the expectation that the difference is often tied to emissions. Because we are interested in the difference between the urban and regional trends (i.e., urban-region), a negative difference indicates only that the urban trends are less than those of the region but does not indicate the sign of the absolute trend. For example, urban X_{CO} decreasing faster than its surrounding region, or a regional increase in X_{CO} with a smaller urban X_{CO} increase would both have a negative difference. First, we follow the general approach of Worden et al. (2012) and use the gridded $0.4^\circ \times 0.4^\circ$ product and define urban grid cells as those where at least 30% of the grid area overlaps with the urban extent. The region is defined as the remaining cells in a $5.2^\circ \times 6.0^\circ$ area centered on the city. We will refer to this as the “gridded” boundaries method. The second method, which we will refer to as the “circular boundaries” method, uses the standard L2 product where monthly averages are computed from all soundings with center points within the urban boundaries, and the region is composed of soundings within a 250-km radius of the city center, excluding the urban area. We include the circular boundaries method as a check on the gridded method but do not expect a

significant difference between them. We use these small areas to try to distinguish how X_{CO} within an urban area is changing, due perhaps to anthropogenic emission changes, independent of broader changes.

For each urban area, we separately calculate the trend within the urban boundaries and the trend for the greater region using the annual averages. For each trend, we find the 95% CI. We then calculate a difference in trends and the corresponding combined CI.

4. Global Analysis

4.1. Impacts of Major Events

Before considering trends, we first consider the impacts of events that emit significant amounts of CO. These irregular events such as biomass burning from wildfires can have significant impacts on CO levels hundreds of kilometers away or more from the source. For example, fires increased carbon release in tropical Asia in 2015 (Liu et al., 2017) which increased CO not only over that region but also was likely responsible for the increased CO over a larger 10,000–20,000 km area. To examine this effect, we use the monthly anomalies (Section 3.1). Results in Figure 1 show this effect of excess CO in the total column affecting a large area. In August 2015, X_{CO} was equal to or slightly elevated above Indonesia compared to the average of all Augusts from 2002 to 2017. In September 2015, we see ΔX_{CO} increased over a larger area. In October and November, the elevated ΔX_{CO} continues to spread in extent, possibly affecting areas 10,000 km away or more. There also appears to be elevated ΔX_{CO} originating from the Amazon during this time. Other major events may have also increased X_{CO} , which prevents a full source location attribution without a model. It is common to have a large irregular event somewhere in the world at least once a year.

While the 2015 Indonesian fires are one of the more striking examples, other regions have also been source locations for major events. Examples of impacts from other major events are shown in Figure 2. During 2003, fires in Siberia burned over 200,000 km² and released around 401–684 Tg C (Huang et al., 2009), including 55–139 TgCO. The area burned in Siberia in the 2003 fires was 3 times the area burned by medium-sized fires since 1995 (Kasischke et al., 2005). Though not as large as 2003, 2002 also had large boreal fires. These large fire years in the early part of the MOPITT record could influence the long-term trends of X_{CO} . Similarly, fires in the Amazon in 2005, 2007, 2010, and 2015, usually around September, increased X_{CO} loading compared to other years. Typically less than 1% of southeastern Amazon forests burn, but in 2007 and 2010, 12% and 5% were burned, respectively (Brando et al., 2014; Deeter et al., 2018). Excess CO from these Amazon fires was transported by Rossby waves southeast across the Atlantic. In 2014, the Northwest Territories complex fire burned 34,000 km² and emitted 164 TgC (Veraverbeke et al., 2017). The burn area was the highest on record for the Northwest Territories going back to 1975 and was about 6 times as large as the average (7.5 times as large as the 2000–2015 average). Column enhancements in CO and other fire tracers at Eureka, NU (80°N) during July and August were attributed to these fires (Lutsch et al., 2016). In terms of area burned in managed Canadian forest since 2000, 2002, and 2014 had the largest burn area (Domke et al., 2018). Our final example is from February 2016, when there were higher than average X_{CO} anomalies over Africa. Africa accounts for 43% of the global 2001–2016 total fires (Earl & Simmonds, 2018). December–February is fire season for equatorial Africa, and while high X_{CO} levels are expected in this region during this time, 2016 was higher than average. This could be linked to the 2015–2016 El Niño, which may have also stressed vegetation in the tropical Rainforest leading to more VOCs being emitted and reacting to form CO.

Major events highlighted here indicate the importance of considering regional changes in CO when examining behavior at a more local scale, such as cities. They also provide examples of how long-term trends could be affected by events that do not occur regularly or at the same location.

4.2. Global Trends

We find over the 16 years of study, there is significant global heterogeneity in the trends (Figure 3). For example, CO columns over most of the NH are decreasing (esp. poleward of 30°N) while the change in the SH is closer to zero. Eastern China is decreasing more strongly than the rest of the NH. The trend over India is mostly neutral. However, an increase in CO emissions from India might be masked by decreasing

Figure 1. Monthly anomalies in X_{CO} during 2015. Here and throughout, we use an Eckert IV equal area projection. Anomalies are the 2015 monthly value minus the average value for the month from 2002 to 2017. In September, we can see the influence of the Indonesian fires on X_{CO} . In October and November, the elevated X_{CO} influences an even larger region, which could overwhelm changes at more local scales due to localized changes in fluxes. Some banding is seen from the individual satellite overpasses.

CO columns over the surrounding areas. Column CO over south central Africa is increasing slightly, which could be due to fires and significant interannual variability. Buchholz et al. (Air pollution trends measured from Terra: CO and AOD over industrial, fire-prone, and background regions, submitted 2021) noted more neutral trends in this region, due perhaps to their use of the thermal IR only product which is more stable but less sensitive to changes closer to the surface.

Figure 2. Monthly anomalies in X_{CO} during times of other major CO-producing events. Large May 2003 ΔX_{CO} in the NH likely largely a result of wildfires in Siberia. September 2007 shows the influence of Amazon fires. During August 2014, there were large wildfires in the Canadian Northwest Territories. February 2016 elevated X_{CO} could be due to larger than usual African fires. NH, Northern Hemisphere.

Figure 3. (a) Average X_{CO} for monthly means during 2002–2017. Regions of elevated X_{CO} (such as west central Africa and eastern China) are noticeable. (b) Overall trend from 2002 to 2017 as a percent change compared with the overall mean. Eastern China has the strongest decrease. Trends over India are within 1σ of zero, but the surrounding region mostly has a negative trend. (c) Standard deviation of the gridded monthly averages. The large standard deviation over the Amazon is primarily due to a large seasonal cycle. (d) Standard deviation of the trend shown in (b) determined from a bootstrap analysis (Section 3.1) and assuming a normal distribution of slopes.

Trends over subsets of 5 years are often dissimilar from trends over the full 16 years of this study. This is perhaps not surprising given the influence of significant events such as the 2002–2003 boreal fires. For X_{CO} , we would also not expect trends in atmospheric loading of CO to continue indefinitely. For example, X_{CO} in a region may decrease due to an increase in newer catalytic converters and the effect of this change will eventually saturate. Similarly, X_{CO} could increase due to more vehicles (esp. motorcycles and scooters) in a region and this effect could also saturate as car ownership per person increases. Piece-wise fitting may make more sense as CO emissions change.

Interestingly, the distribution of CO emissions in EDGAR and high X_{CO} levels as seen by MOPITT do not always line up (Figures 3a and 4a). While both agree on large amounts of CO over the Eastern United States, eastern China, and India, MOPITT observations also show elevated CO from Africa likely from VOCs. CO production from VOCs is not accounted for in the inventories.

Figure 4. Maps of gridded inventory (EDGAR 4.3.2) averages and trends over 2002–2012. (a) Average CO emissions show high emission regions especially in China and India with other hotspots throughout the world, while the US emissions are more dispersed. (b) Trends in CO emissions show a large decrease from the United States. Europe and Japan, with mixed changes from China. India, especially northern India, shows a large increase in emissions over the time period. EDGAR, Emissions Database for Global Atmospheric Research.

Figure 5. Trends in X_{CO} for different 5-year windows. A few features for each map are highlighted here. (a) Trends for 2002–2006 characterized by strong decreases above 45°N. (b) NH trends for 2005–2009 are weaker overall, with strong decreases over eastern China and southeast Asia. (c) During 2009–2013, the rate of decrease in the NH continued to decline, and the SH slightly increased in X_{CO} . (d) The rate of decrease for China is similar to the rest of the NH for 2013–2017. White areas indicate no trend or in some areas (especially close to the poles) insufficient data. NH, Northern Hemisphere; SH, Southern Hemisphere.

There are also significant differences in the trends shown by EDGAR and MOPITT (Figures 3b and 4b). EDGAR shows a strong decrease over the United States, which is observed by MOPITT. However, the strong decrease over eastern China is not as apparent in the EDGAR inventory. India also has an increase in the inventory compared with a more neutral change seen by MOPITT which could be due to a local increase coupled with a larger regional decrease.

Trends for four different overlapping 5-year time windows show significant variability (Figure 5). Over 2002–2006, the strong decrease above 45°N is likely related to unusually large boreal forest fires during 2002 and 2003. Significant decreases in eastern China are likely related to reduced fossil fuel emissions of CO. In general, the SH appears to be slightly increasing. During 2005–2009, the NH is still decreasing, though not as strongly as in 2002–2006. X_{CO} over eastern China continues to decline through 2009–2013, but the rate of decrease slows and becomes similar to the surrounding region during 2013–2017. Parts of the NH increased in X_{CO} during 2009–2013. Increasing X_{CO} levels for 2009–2013 as well as 2013–2017 are seen in south central Africa and, to a lesser extent, the Amazon. These may be related to the overall slightly increasing SH trend. The 2013–2017 timeframe includes the 2015–2016 El Niño, which had regionally specific carbon cycle responses. Liu et al. (2017) found increased CO₂ emissions due to increased fires over SE Asia, higher temperatures and more respiration over Africa, and reduced gross primary productivity over South America due to drought. From these plots, it appears that the sharp declines in X_{CO} seen closer to the beginning of the century are beginning to level off.

Because of the increase in CO₂ from China and SE Asia (Friedlingstein et al., 2019), the decrease in X_{CO} may seem counterintuitive. However, the decreasing CO since the early 2000s in this region has already been noted (Jiang et al., 2017; Worden et al., 2013; Yin et al., 2015; Zheng et al., 2018). Studies agree that the decrease is due to anthropogenic source reduction. The seemingly incompatible increase in CO₂ with a decrease in CO emissions is reconciled by considering technology changes which have led to decreased CO emission factors. R. Wang et al. (2014) also noted that emission factors or emission intensities for black carbon have decreased significantly for China and India over recent decades.

Figure 6. Trends in X_{CO} for seasons during the 2002–2006 period: (a) December–February, (b) March–May, (c) June–August, and (d) September–November.

We also find large variability when fitting seasons within a given 5-year span (e.g., 2002–2006, Figure 6). The sign of the trends can differ depending on season, for example, over Africa and in the western Pacific. In general though, trends are similar over broad thousand kilometer regions or larger. We find the SON season to have the strongest negative NH trend over 2002–2006, likely from large fires mostly in Siberia in 2002 and 2003. Larger than average boreal fires in 2002 had the most influence on X_{CO} during SON. Eastern China appears to have an overall negative trend across all seasons. The trend over the SH oceans is more positive in DJF than in SON. These broad trends highlight how regional variability needs to be understood when trying to understand trends at smaller scales on the order of $40 \text{ km} \times 40 \text{ km}$.

There are 22 TransCom regions including 11 regions over land and 11 over water (Gurney et al., 2003). A map of these regions is in the Figure S1. For each of these regions, we have calculated the average trend during 2002–2017 and over the four 5-year subsets (Table 2, and Figure 7). Over 2002–2017, long-term trends are between 0.1 and $-0.9 \text{ ppb year}^{-1}$. For the 5-year subsets, there are wider dynamic ranges from -3.6 (Eurasia boreal) to 0.6 (Southern Africa) during 2002–2006 and -1.5 (Europe/Eurasia boreal) to $1.8 \text{ ppb year}^{-1}$ (Southern Africa/Southern America Tropical) during 2013–2017. This again shows not only the spatial variability but also the temporal variability in X_{CO} trends. For example, the pairwise difference between the two 5-year subsets ranges from 2.1 (boreal, indicating a slowdown of decreasing CO) to $-0.8 \text{ ppb year}^{-1}$ (Tropical Asia, indicating an acceleration in the CO decrease). Together, these indicate that subsets of the longer time series need to be considered to better understand trends. Broad patterns among the TransCom regions can be seen in Figure 7. We again see a general decline in the NH regions, which has weakened in more recent years. In the SH, regions have gone from no trend, to a slight increase during the 2009–2013 and 2013–2017 time periods.

Various measurement uncertainties could make spurious trends appear in a data set including trends in the a priori profiles, trends in noise, and trends in AKs. Compared to measurements from various TCCON sites, there is a small negative trend of $-0.06\% \text{ year}^{-1}$ or about $-0.05 \text{ ppb year}^{-1}$ in the MOPITT observations (Hedelius et al., 2019). For the case of MOPITT V7J, the a priori is the same for different years (Deeter et al., 2014). Though the a priori will not directly contribute to interannual trends, it can contribute indirectly likely as a dampening effect as the true atmosphere moves away from the prior combined with the AKs. Example column AKs are in Figure 8. Large measurement errors could lead to low degrees of freedom and cause values closer to the prior to be returned, decreasing any inferred trends. MOPITT instrument noise

Table 2

Average Trends and Standard Deviations in TransCom Regions in ppb Year⁻¹

Region	2002–2017	2002–2006	2013–2017
North American boreal	−0.9 ± 0.3	−3.3 ± 1.5	−1.2 ± 1.2
North American temperate	−0.8 ± 0.2	−2.3 ± 1.0	−1.1 ± 0.9
South American tropical	−0.3 ± 0.2	0.5 ± 1.9	1.8 ± 1.9
South American temperate	−0.2 ± 0.1	0.1 ± 0.8	0.9 ± 0.7
Northern Africa	−0.5 ± 0.3	−1.3 ± 1.0	−0.4 ± 0.8
Southern Africa	0.1 ± 0.3	0.6 ± 1.6	1.8 ± 1.4
Eurasia boreal	−0.8 ± 0.2	−3.6 ± 1.5	−1.5 ± 1.3
Eurasia temperate	−0.7 ± 0.4	−1.8 ± 1.4	−0.9 ± 1.0
Tropical Asia	−0.8 ± 0.6	−0.4 ± 1.9	−1.2 ± 1.9
Australia	−0.2 ± 0.1	0.1 ± 0.5	0.5 ± 0.3
Europe	−0.9 ± 0.2	−3.2 ± 1.2	−1.5 ± 0.9
North Pacific temperate	−0.8 ± 0.2	−1.9 ± 1.2	−1.3 ± 0.9
West Pacific tropical	−0.3 ± 0.2	−0.3 ± 0.6	−0.7 ± 0.9
East Pacific tropical	−0.3 ± 0.1	−0.4 ± 0.6	−0.1 ± 0.4
South Pacific temperate	−0.1 ± 0.1	0.3 ± 0.4	0.7 ± 0.4
Northern Ocean	−0.7 ± 0.3	−2.3 ± 1.3	−1.3 ± 1.2
North Atlantic temperate	−0.7 ± 0.2	−1.5 ± 0.8	−0.9 ± 0.6
Atlantic tropical	−0.2 ± 0.1	−0.4 ± 0.6	0.5 ± 0.9
South Atlantic temperate	−0.2 ± 0.1	0.0 ± 0.6	0.7 ± 0.4
Southern Ocean	−0.1 ± 0.1	0.2 ± 0.8	0.7 ± 0.7
Indian TROPICAL	−0.2 ± 0.2	−0.2 ± 0.6	−0.4 ± 1.0
South Indian temperate	−0.1 ± 0.1	0.4 ± 0.5	0.9 ± 0.4
Northern Hemisphere	−0.7 ± 0.3	−1.8 ± 1.5	−1.0 ± 1.0
Southern Hemisphere	−0.1 ± 0.1	0.2 ± 0.8	0.7 ± 0.8
Global	−0.4 ± 0.4	−0.9 ± 1.6	−0.2 ± 1.3

Note. Trends where one standard deviation does not cross zero are in bold. See also Figure 7.

has been slowly increasing (Deeter et al., 2015). We quantify the dampening effect of the anchoring to a prior by calculating the ratio of the expected change with and without the AK:

$$R_d = \frac{\log_{10}(d+1) \sum a_i}{c_a d} \quad (1)$$

where c_a is the column prior, d is the relative change based on the gridded 2002–2017 slopes at the time of the measurement, and $a_i = \frac{\partial \hat{c}}{\partial \log_{10} x_i}$ are AK elements (Hedelius et al., 2019). We find a mean ratio of 0.90 (95% CI: 0.63, 1.26), indicating an average dampening effect of 10% due to being tied to the prior. There is a weak negative relationship ($r = -0.32$) with time, where the ratio is changing by -0.003 per year, indicating an increasing reliance on the prior.

Trends in noise may also lead to AK trends. We have also not accounted for how changing AKs could make a trend in the retrieved X_{CO} , which adds uncertainty (Yoon et al., 2013). As an uncertainty estimate, we examine trends in column AKs over temperate North America and find drifts from -2.5% year⁻¹ to 1.9% year⁻¹ depending on the layer. The lowest six layers have a negative trend, while the upper four layers have a positive trend and the column average is -0.7% year⁻¹. The effect on retrieved column CO depends on the difference between the true atmospheric state and the prior CO profiles. A consistent +20 ppb difference

Figure 7. Trends for TransCom regions for different time subsets. These are arranged approximately from north to south. Blue labels are for water regions, brown labels are for land regions, and gray labels are for larger mixed regions. Error bars are standard deviations of the trends among individual $0.4^\circ \times 0.4^\circ$ grid cells across the region. Magenta symbols are trends derived from the EDGAR 4.3.2 inventory.

from the a priori profile throughout the column would lead to about a -0.06 ppb year $^{-1}$ trend in X_{CO} . Jiang et al. (2017) showed significantly different MOPITT AKs for 2000, which we have excluded here along with 2001 measurements due to the different instrument configuration.

Figure 8. Examples of mean column AKs across select years that fall within the boundaries of select cities. Shaded regions go from the 10th to 90th percentiles. Matrix AKs were converted to column AKs following the methods of Hedelius et al. (2019). These generally indicate higher but decreasing sensitivity in the lower troposphere. AK, averaging kernel.

Unlike some other studies, we have not attempted to separate out different contributions to CO trends (Jiang et al., 2017). A study by Turner et al. (2017) suggested a decrease in the OH radical since around 2000, though results by Gaubert et al. (2017) showed no large interannual variability. Because of the coupled CO–OH–CH₄ system, a decrease in CO emissions leads to an increase in CO chemical production from VOC oxidation (Gaubert et al., 2017). Jiang et al. (2015) showed large variability in CO emissions depending on prescribed OH fields, though Gaubert et al. (2017) showed that global OH concentrations are well buffered. Studies tend to agree that a change in OH is not the cause of decreased CO this century (Jiang et al., 2017).

5. Trends in Selected Urban Areas

Measurement uncertainty, spatial smoothing, regional transport, and proximal cities may obfuscate trends in X_{CO} from a single urban area. MOPITT footprints are ~484 km², which is larger than the median area (335 km²) of the 500 most populated cities in this study. Though a city impacts the atmosphere beyond its borders, the MOPITT spatial footprint and measurement uncertainty lead to uncertainty in urban X_{CO} trend analysis. Places like India and eastern China are known to have high urban density, but even in the rest of the world the median distance between the closest urban centers is less than 30 km. This is likely skewed toward suburban areas clustering around larger cities. Because cities in a region will likely have similar behavior in emission changes (e.g., changing type and density of vehicles), we could expect regional X_{CO} trends in some cases to be related to emission changes originating from cities. Trends in urban areas are linked to nearby cities not only because regional trends can overwhelm urban trends but also because the urban emissions themselves can affect the regional trends, especially in highly populated areas.

We first find cases where differences in trends are all negative, that is, X_{CO} is decreasing faster in the city than the region. The 95% CI in the difference does not cross zero and is negative in 9 and 15 urban areas (of 500 total) for the gridded and circular boundaries methods, respectively. The intersection of these sets includes just three urban areas: Guangzhou (China), Mexico City (Mexico), and Manchester (England). Both methods have three areas with positive trends where the 95% CI does not cross zero, but the three areas are different for each method.

The relatively few number of cities with significant trends leads to the question: why are there not more? Emission inventories show large changes in CO emissions (e.g., Figure 4) and there are large regional X_{CO} changes observed in the MOPITT data (Figure 3). We explore this with two case studies. The first is Los Angeles (LA) which is known for historically poor air quality. The next is Mexico City which, like LA, is a large basin city that is relatively isolated from other urban areas. These factors make them best case scenarios for observing a change.

5.1. Case Studies

Ideally, we would compare satellite-derived trends with trends measured from the ground. However, there is a paucity of long-term (decadal or longer) ground-based column measurements within urban areas. One of the exceptions to this is LA where there have been observations made from the MkIV spectrometer since the mid-1980s. In the early 1950s, LA CO emissions were around 1,800 GgCO year⁻¹ from incomplete combustion which, combined with uncombusted VOCs was a 10%–15% loss in fuel combustion value (Haagen-Smit, 1970), not to mention a precursor to health issues. Emissions rose to around 5,100 GgCO year⁻¹ in 1968, and a goal was set for an 85% reduction in CO emissions from new cars starting in 1975—the same year the catalytic converter was introduced. Despite an increase in the number of vehicles, CO emissions are now around 500 GgCO year⁻¹ (Hedelius et al., 2018).

A decrease in atmospheric X_{CO} in LA since the mid-1980s can be seen in Figure 9. The seasonal averages of the MkIV measurements are composed of about 5–15 days of measurements each, with as few as 1 and as many as 26. Seasonal averages within the basin show significant scatter in the early record, due to large day-to-day variability from variation in ventilation patterns combined with a large flux. Through the mid-1990s, values above 200 ppb were not uncommon within the basin. Since about 2009, a little over half of the measurements have been less than 100 ppb. The differences in the lower panel are subject to uncertainty because we have not made adjustments for the influences of AKs or different measurement altitudes. Altitude differences can lead to different column amounts of gases that are not vertically uniform (Hedelius

Figure 9. Upper panel: seasonal averages of X_{CO} measurements from ground-based instruments compared with measurements from MOPITT within the Los Angeles basin and surrounding region. A decrease within the city can be seen from the mid-1980s to present. Different triangle directions for the MkIV indicate different measurement locations: JPL (345 m, orange down arrow), Ft. Sumner, New Mexico (1,260 m, right arrow), Daggett, California (626 m, up/right arrow), and Table Mountain, Wrightwood, California (2,260 m, left arrow). Error bars represent standard errors of the mean for MkIV observations. The dashed line in the upper panel is the least squares fit to the MkIV regional values. Labels ci, jc, jf, and df are for different TCCON sites (Section 2.2). Lower panel: differences of city minus region (or background) are shown without accounting for differences in averaging kernels or surface altitudes and hence are subject to some uncertainty. MkIV - MkIV differences are city minus the fitted line because regional observations were made at different times from city observations. MOPITT, Measurements Of Pollution In The Troposphere; JPL, Jet Propulsion Laboratory; TCCON, Total Carbon Column Observing Network.

et al., 2017). Taking the basic differences, enhancements (MkIV - MkIV) in the basin were around 100 ppb compared to the surrounding region through the mid-1990s. MOPITT - MOPITT enhancements shown in the lower panel are from the circular boundary method and the averages are 10 ppb from 2002 to 2008 and 4 ppb from 2009 to 2017. The average TCCON - TCCON difference is around 10 ppb, which may be larger than the MOPITT estimates because the TCCON instruments measure the atmosphere closer to the urban center. TCCON instruments also measure throughout the entire day, whereas MOPITT only measures in the morning when the basin is likely less polluted. The large but declining X_{CO} through the 1990s show that much of the decrease in historically high CO levels occurred before the MOPITT record begins. This helps explain why the trend as observed by MOPITT is not as large in LA and possibly other locations. For the remaining global urban areas, we only include MOPITT observations.

Trends for Mexico City are shown in Figure 10. For this case, we see a wide variability in regional gridded X_{CO} (~20 ppb) each month, with the urban area nearly always in the top 95th percentile. Overall, the regional X_{CO} average is decreasing, but X_{CO} within the urban area is decreasing faster. From the maps, we see the population density and urban boundaries are constrained to the center grid cell due in part to the surrounding mountains. The topography traps CO and other pollutants. Though median X_{CO} levels are still enhanced by ~15–20 ppb over the urban region, there has been a decrease in the enhancement (Figure 10c). This study shows that longer term changes may be difficult to observe because of significant seasonal variability along with years that appear anomalous (e.g., 2004).

5.2. Global Patterns in Urban Areas

We also include examples for several other urban areas in the Figures S2–S12. For the most part, the cities we examine are located in regions with decreasing X_{CO} . On top of these regional X_{CO} changes, there may be urban changes which are often difficult to distinguish. The 95% CI of the difference between the urban and regional trends nearly always crosses zero (e.g., Figures 11 and S13–S16). Thus, the trends within the urban areas tend to change in concert with the region at large. CIs shown in Figure 11 are large, highlighting large uncertainties in trends using these data and methods. These large uncertainties may be from

Figure 10. Trends of MOPITT X_{CO} over Mexico City. (a) Colored points represent monthly averages of grid cells within the urban area as noted in (d). Gray heatmap is for monthly averages from all grid cells in full region. (b) Solid lines are averages of urban and nonurban grid cells. Red fill indicates higher urban X_{CO} , and blue fill indicates higher surrounding regional X_{CO} . Percentiles of urban grids compared with those throughout the full region are shown as dots. (c) Gridded monthly differences correspond to the difference between solid lines in (b). Area monthly averages are averages within shown urban boundaries (gray borders in maps) compared to the average outside of urban boundaries, but within 250 km of the center. Annual averages also shown. (d) Median X_{CO} of the monthly averages. Red star marks the urban center. Purple triangles mark grid cells that define the urban area (partially obstructed by the red star here). Gray line indicates the urban boundaries. Cyan dots mark other urban areas with a population of at least 50,000. A Lambert azimuthal equal area projection is used. (e) Same as (d), but showing the trend within each grid cell from Figure 3b. (f) Map of population density, gridded to 0.1° × 0.1°. MOPITT, Measurements Of Pollution In The Troposphere.

some combination of sensor noise, retrieval limitations, or from actual large variability in X_{CO} compared to trends. For example, Jakarta has a large uncertainty which is likely due to the influence of large biomass burning events that differ significantly in magnitude from year to year (e.g., see Figure 1). There is a lack of readily available information on Mbuji-Mayi, but it appears to be a developing city that likely in part uses nonoptimized combustion. Either limitations on our methodology or the developing status of Mbuji-Mayi combined with a population change from about 1.9 to 3.0 million from 2000 to 2015 could explain the increase in CO columns.

Though a slight majority (53%) of the cities are decreasing in X_{CO} relative to the regions, this is not significant using a two-tailed binomial distribution test ($p = 0.19$). This analysis is not able to support or oppose the findings by others that the global decline in CO is due in part to reductions in anthropogenic CO emissions (Jiang et al., 2017; Yin et al., 2015). Ignoring sensor limitations, such as noise and footprint sizes, even if urban emissions are changing it may be difficult to distinguish them from their surrounding regions because cities themselves could be driving the regional changes on these time scales. If CO can mix throughout a region on a time scale of a few days, then “upwind” cities may be causing the regional changes.

The CO enhancement compared to the region (ΔX_{CO}) is persistently greater than zero in many cities, but it may not be changing from year to year. This indicates a locally polluted atmosphere that is not getting better or worse.

Figure 11. Difference in trends in annual averages between city and surrounding region from 2002 to 2017 using the 250-km region method. Error bars represent the 95% CI from a bootstrap analysis ($n = 600$). The 20 most populous urban areas are shown here along with 10 others we selected, sorted by decreasing trend. CI, confidence interval.

Though ΔX_{CO} may be correlated with developed/developing status (Silva & Arellano, 2017; Silva et al., 2013), we postulate that it may be more indicative of local preferences or needs for transportation, heating, and other CO-producing activities. For example, India and Mexico may both be classified as newly industrialized countries, but in India around 80% of vehicles are motorcycles (<https://www.ceicdata.com/en/india/number-of-registered-motor-vehicles/registered-motor-vehicles-all-states>, last access January 25, 2020) with higher CO emissions whereas in Mexico only around 10% of vehicles are motorcycles (<https://www.in-egi.org.mx/sistemas/olap/proyectos/bd/continuas/transporte/vehiculos.asp>, last access January 25, 2020). Over Mexico City, ΔX_{CO} has been decreasing (i.e., the urban trend is more negative than the surrounding region) which could be because Mexico City is more isolated from the region due to the topography. However, ΔX_{CO} has mostly remained constant over Mumbai and Delhi (i.e., the urban trend has been about the same as the surrounding region). Similarly, Paris and New York City are both developed, but X_{CO} enhancements differ over the two cities, possibly due to different preferences in transportation. ΔX_{CO} is around 5 ppb in New York City, compared to around 2 ppb in Paris where a greater percentage of vehicles use diesel fuel. There are cases however in which CO-producing activities may be related with developing status—for example, wood fires for heating.

6. Discussion and Conclusions

Though overall there has been a global decrease in X_{CO} during the MOPITT record, it has not been uniform across regions (Worden et al., 2013). The MOPITT record is long enough that trends should be considered differently on sub-mission time scales. Eastern China, which had some of the highest X_{CO} levels in the early 2000s, has had one of the largest decreases. The rate of decrease has appeared to slow down in the NH recently from -1.8 to -1.0 ppb year $^{-1}$, and X_{CO} in the SH has begun to increase slightly from 0.2 to 0.7 ppb year $^{-1}$. In agreement with other studies, this is likely indicative of diminishing returns from decreasing CO emission factors from gasoline automobiles, which have decreased by more than 10-fold since the 1980s (Jiang et al., 2018; Yin et al., 2015). One hypothesis is the slowdown to slight increase in X_{CO} in recent years could also be due to increasing CO emissions from fires and VOCs related to changing climate. Though biomass burning emissions have been decreasing globally, there is an increasing trend in the boreal region in the Global Fire Emissions Database during 1999–2014 (Arora & Melton, 2018).

We have also seen that large events can have far reaching impacts. It is not uncommon for CO from wildfires to affect regions thousands of kilometers away. This could affect the trend analysis, especially due to large boreal fires in the early part of the record.

Trends in the EDGAR CO emissions inventory and atmospheric trends observed by MOPITT do not always match (Figures 3b and 4b). This is likely due to a combination of factors including mixing in the atmosphere, unrepresentative emission factors in the inventory, CO production by VOCs, and unaccounted for changes in sources with large emissions such as wildfires. This demonstrates the continued need for both observations and emission inventories to understand changes in global CO.

Originally, we anticipated determining unique X_{CO} behavior for various cities. We find however that in all but a few cases the cities behave similarly to the surrounding region, which makes distinction difficult. There are several reasons cities may usually not be distinguishable. (1) CO emissions within a city may actually be changing at the same rate as emissions in the surrounding region. Given the clustering of urban areas, this could make regional effects the same as those within a city. (2) AKs may play some role since they peak around the midtroposphere and are smaller at the surface. Thus, retrieved X_{CO} trends here are most reflective of free troposphere trends. This could lead to underamplification of CO changes in the retrievals at the surface compared to the rest of the troposphere. Pollutants emitted at the surface are expected to mix upwards as they spread throughout a larger region. (3) Measurement uncertainty, especially bias in the MOPITT measurements, could affect differences. For example, aerosols are nearly always a source of uncertainty in trace gas retrievals and can be coemitted with CO from urban sources. When compared with individual TCCON sites, MOPITT X_{CO} bias typically varies from 3 to 10 ppb (Hedelius et al., 2019). (4) MOPITT has an individual footprint size of $22 \times 22 \text{ km}^2$ which is on order of the size of cities. The TROPospheric Monitoring Instrument (TROPOMI) launched in 2017 has spatial footprints about 10% of the size of MOPITT and is even better suited to study urban areas (Borsdorff et al., 2018). The Geostationary Carbon Cycle Observatory (GeoCARB) expected to launch in 2022 will have footprint sizes about one third those of TROPOMI and will also be able to make multiple measurements over an area in 1 day (Moore et al., 2018).

Trends in X_{CO} are generally indistinguishable from those in the surrounding regions for this period. However, for some cities, X_{CO} may have decreased significantly before MOPITT observations began. Los Angeles is the only location where we have ground-based column CO measurements going back to the 1980s, but this case study highlights the positive effects of technological improvements and emissions regulations. In the 1960s, Los Angeles was the smog capital of the world (Haagen-Smit, 1970), but now X_{CO} levels are at or near those of the rest of the surrounding region. Smog mitigation efforts that started in LA have been adopted in much of the rest of the world leading to much better air quality and more efficient vehicles. This transition to cleaner combustion, along with large regional specific declines in X_{CO} , shows the positive impact of a single city or region.

Conflict of Interest

The Authors declare no conflict of interest.

Data Availability Statement

MOPITT data may be obtained from the NASA Earthdata website (<https://search.earthdata.nasa.gov/search/granules?p=C1288777617-LARC>, last access: October 30, 2020). TCCON data were obtained through the TCCON data archive hosted by CaltechData (<https://tccondata.org/>, last access: June 19, 2019). Ground-based MkIV data were obtained through the JPL MkIV website (<https://mark4sun.jpl.nasa.gov>, last access: June 19, 2019).

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