Research Paper

Scaling relationship between CO pollution and population size over major US metropolitan statistical areas

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ABSTRACT

As the world's population is projected to reach 9.7 billion by 2050, large and small cities will continue to expand. There are few studies investigating how the size of cities affects air pollution. Carbon monoxide (CO), a precursor of ozone and a by-product of incomplete combustion, is a common air pollutant. The major sources of CO in the US urban areas are motor vehicles. Here we examined the scaling relation of CO concentrations over major US metropolitan statistical areas (MSAs) using Measurement Of Pollution in The Troposphere (MOPITT) surface CO retrievals and National Emissions Inventory (NEI) data. We found significant power-law scaling relationships between CO and population (r^2 of 0.30 for MOPITT average CO concentration and r^2 of 0.71 for NEI total CO emission). We found decreasing CO trends from 2000 to 2015 using MOPITT and EPA CO ground measurements. Sublinear scaling relationships (scaling coefficient $\beta < 1$) suggest that larger MSAs are more combustion-efficient in terms of CO emissions. We found a weaker scaling relation and smaller scaling coefficient from MOPITT CO concentrations than from NEI total CO emission data. This pattern may be attributed to the differences between the two CO datasets: annual average of monthly MOPITT CO concentration at 1° by 1° spatial resolution versus the NEI annual CO emissions compiled from emission inventories and estimated from mobile source emissions models. Future research is needed to investigate the capability of using satellite observations to study scaling relations between air pollutants and population.

1. Introduction

The world population is projected to reach 9.7 billion by 2050 (United Nations, 2015). The majority of the population growth will be absorbed by urban areas. Cities consume materials and energy to support their functioning, and generate waste streams in multiple forms, including air, water, noise, heat, and solids. In analogy of a city as a biological system, Bettencourt, Lobo, Helbing, Kuhnert, and West (2007) found that many urban properties follow power-law scaling relationships with city size, often measured by city's population. Power-law scaling relationships are described mathematically as $Y = Y_0 N^\beta$, where Y represents an urban property, N is population, Y_0 is the intercept, and β is the scaling coefficient. The scaling relations can be grouped into three categories across various urban properties (Bettencourt et al., 2007; Bettencourt, 2013): a superlinear scaling with $\beta > 1$ has been observed in socioeconomic activities (such as wages

and patents), suggesting that larger cities generate wealth and creativity out of proportion to their size. Sublinear scaling with $\beta < 1$ has been found in urban infrastructure and material quantities (such as roads), indicating per capita measurement decreases with city size due to more efficient use of infrastructure and materials; and linear scaling with $\beta \approx 1$ reflects a proportional increase in urban properties with city size, such as water consumption.

A few studies have investigated whether larger cities are more CO_2 emissions efficient than smaller cities. Fragkias, Lobo, Strumsky, and Seto (2013) reported a linear scaling relation between CO_2 emissions (measured in metric tons) and population over the urban 'core based statistical areas' (CBSAs) of the US. In contrast, Oliveira, Andrade, and Makse (2014) found a superlinear scaling across US cities using the same *Vulcan Project* data. They ascribed the discrepancy to the different definitions of city boundaries: a linear scaling for metropolitan statistical areas (MSAs) and a superlinear scaling for cities defined by

connected urban settlements (Oliveira et al., 2014). A more recent study concluded that the scaling of CO_2 emissions (tons) and population depends on the country's economic development: superlinear scaling for cities in economically less developed countries and sublinear scaling for cities in developed countries (Rybski et al., 2016). While a mixed result reported for CO_2 emissions, very few studies investigated the scaling relationships between air pollutant concentrations and city size. Lamsal, Martin, Parrish, and Krotkov (2013) used satellite measurements to study the relationship between NO_2 and urban population using a systematical sampling approach, and reported a sublinear relationship. Han, Zhou, Pickett, Li, and Li (2016) utilized the Global Annual $PM_{2.5}$ dataset to investigate the relationship between $PM_{2.5}$ and urban population size.

While previous research has investigated the relationship between two common air pollutants (i.e., NO_2 and $PM_{2.5}$) and urban population (Han et al., 2016; Lamsal et al., 2013), the relationship between other common air pollutants and population size is unclear. Here we study the relationship between population size and carbon monoxide (CO) using satellite measurements and National Emission Inventory (NEI) data to provide another perspective on the relationship between urban size and air pollution. In urban areas, CO is primarily formed by incomplete combustion of carbon-containing fuels, and mobile sources (including onroad vehicles and off-road mobile sources) are the biggest contributor to total CO emissions in the US (EPA, 2011). Thus, CO can be viewed as a waste product of energy consumption used for movements supporting the functioning of cities. The level of CO concentrations in a US city is largely associated with fuel efficiency of vehicles, the degree of commuting and the distance to suburbs, the degree of public transportation, as well as traffic conditions. In the US, private automobile is the dominant travel mode ($> 80\%$) (McKenzie, 2015). Although the rate of automobile commuting declines from 87.9% to 85.8% from 2000 to 2013, the number of workers who commuted by private vehicle increased continuously from 1960 to 2009 (McKenzie & Rapino, 2011; McKenzie, 2015). Average travel time for workers was about 25 min in 2010 and remained the same in 2009 (McKenzie & Rapino, 2011). We hypothesized that urban CO concentrations follow a power-law scaling relationship with population size. We examined the scaling relationship using both satellite CO measurements and NEI CO data. The comparison of the scaling relationship resulting from the two CO datasets can reveal the capability of using satellite CO to study scaling relations between CO and population. In addition, we also examined changes of CO level over major US cities using non-parametric trend analysis on both satellite CO and ground measurements over the same time period.

2. Data

We used CO concentration data from Measurement Of Pollution in The Troposphere (MOPITT), the National Emissions Inventory (NEI), and EPA air quality sensors. While MOPITT provides 1 by 1° grid observations, NEI CO data are available at the county level. Temporal coverage of MOPITT CO data in this study runs from April 2000 to December 2015. The June and July 2001, and the August and September 2009 MOPITT data are missing due to a halt in instrument operations. Corresponding to MOPITT's temporal coverage, we selected NEI data at a three-year interval for analysis: 2002, 2005, 2008, 2011, and 2014. Both MOPITT and NEI CO data were used to build relationships with city's population. MOPITT and *in-situ* EPA CO data were used to study trends.

2.1. MOPITT CO

MOPITT is a spectrometer with thermal infrared (TIR) and near-infrared (NIR) channels on board the Terra satellite that has local time descending node equatorial crossing around 10:30. The instrument is designed to detect atmospheric CO absorption around 4.7 μm (TIR

channels) and 2.3 μm (NIR channels). It has a nominal horizontal spatial resolution of 22 by 22 km at nadir. The MOPITT V3 retrieval products, released in 2000, were the first satellite dataset for global troposphere CO (Deeter, 2013).

We used the monthly MOPITT V6 L3 data product, which is the spatial average calculated by projecting L2 data onto a 1° grid. Retrieval errors associated with random instrument noise are reduced in the monthly L3 data compared to other MOPITT data products because the monthly L3 data are averaged both spatially and temporally. The TIR-only product has the highest temporal stability with relative lower retrieval errors and bias drift compared to the TIR + NIR product (Deeter et al., 2014). Validation using *in situ* data from NOAA validation sites, the V6 TIR-only products exhibited minimal negative bias drift of $-0.16 \pm 0.11\%/y$ at the surface (Deeter et al., 2014). MOPITT sensitivity to surface CO varies geographically. Surface-level CO sensitivity not only strongly depends on the thermal contrast between surface and the lower atmosphere, but is also affected by geophysical noise that is relatively strong over terrain with high relief or topographic complexity. The MOPITT averaging kernel (AK) matrix, which calculated from weighting functions, *a priori* covariance matrix, and instrument error covariance matrix, describes the sensitivity of the retrieved profile to the true profile (Deeter et al., 2014). The first or second element of the diagonal of the AK matrix reveals MOPITT sensitivity to surface CO at 1000 hPa or 900 hPa respectively (for cities at higher elevation, such as Denver, their surface level CO is at 900 hPa). A low AK value suggests poor measurement sensitivity to CO and the resulting CO estimate is dominated by the *a priori* profile. Therefore, we used the first or second element of the diagonal of the AK matrix to identify high quality surface CO observations and then selected MSAs with good retrieval sensitivity. This study used an AK threshold of 0.1 to identify high quality MOPITT surface CO observations (Worden et al., 2010).

2.2. The NEI and in situ CO

The NEI provides a detailed estimate of emissions of criteria pollutants, criteria precursors, and hazardous air pollutants (EPA, 2017). The US EPA releases NEI every three years based primarily upon data provided by State, Local, and Tribal air agencies for sources in their jurisdictions and supplemented by data developed by the EPA (EPA, 2017). Sources of emissions estimates are grouped into five categories: point sources, nonpoint sources, onroad sources, nonroad sources, and 'event' sources (EPA, 2017). Emissions from onroad and nonroad sources are estimated from mobile source emissions models (EPA, 2017). The NEI data are available at three summary levels: Tier 1, Tier 2, and Tier 3. The NEI Tier 1 summaries provide emission data for 14 Tier 1 categories, including three categories in fuel combustion (electric utility, industrial, and other), chemical & allied product manufacturing, metals processing, petroleum & related industries, other industrial processes, solvent utilization, storage & transport, waste disposal & recycling, highway vehicles, off-highway, miscellaneous, natural resources, and biogenic emissions (EPA, 2017). In addition to total CO emissions, we aggregated county-level highway vehicle emissions for each MSA, because highway vehicle CO emissions are the biggest contributor to total CO emissions ($> 80\%$) in US cities. We selected all available EPA ground monitoring sites in each MSA with CO records covering 2000–2015, the same temporal coverage with the satellite data. The number of ground monitoring sites in each MSA varies from one to thirteen. *In situ* CO data are in the form of an eight-hour average. We first averaged the highest CO value for the day from the selected monitoring sites by month, and then spatially averaged multiple sites to represent the CO level of a specific MSA.

2.3. Population data

We used LandScan global population distribution data at 1 km resolution (Bright, Rose, & Urban, 2013) to estimate total population
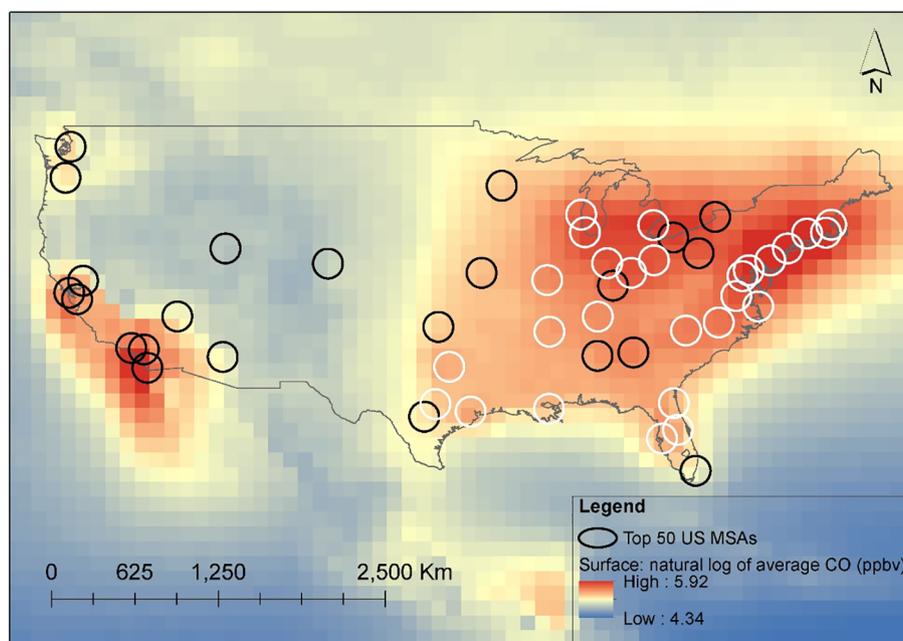
Fig. 1. Top 50 US metropolitan statistical areas (MSAs) with the selected 27 MSAs highlighted in white: 1° buffer of each urban center superimposed on natural log of long-term average MOPITT surface CO.

within each 1° buffer area and used county-level US Census population estimates to calculate total population for each MSA. The LandScan population data represent ambient population or average population distribution over a 24-hour period (Bhaduri, Bright, Coleman, & Urban, 2007). Annual population data were available to us from 2000 to 2015, with the exception of 2011.

3. Methods

3.1. Sampling and quality control of MOPITT data

Due to the coarse spatial resolution of a 1° grid cell of MOPITT data, each grid cell only covers a portion of its nominal MSA area. Therefore, we applied a 1° buffer to each urban center for consistent sampling of MOPITT surface CO retrievals and population (Fig. 1). Each 1° buffer area overlaps with multiple MOPITT grid cells. We excluded MOPITT grid cells covered by 50% or more oceans or lakes because of low retrieval sensitivity to surface CO over water surface (Deeter, Edwards, Gille, & Drummond, 2007). We used a weighted average approach to extract MOPITT data for each 1° buffer: within each 1° buffer, we first calculated the percent area for each overlapping area between 1° buffer and MOPITT grid cells, and then multiplied this by its corresponding MOPITT grid cell value of CO retrievals. We calculated annual average CO from monthly CO retrievals for each city. Due to inherent limitations of MOPITT sensor, we used AK values that come with the MOPITT CO data products to select high quality surface CO observations. We applied the same weighted average approach to extract AK values. To determine overall MOPITT sensitivity to surface CO over a city, we calculated the 15-year mean of MOPITT AK values by averaging the data of the entire study period. Cities with AK mean of < 0.1 were deemed as locations where MOPITT exhibited overall poor sensitivity to surface CO. Among the top 50 MSAs in the US, 23 MSAs had an average AK of < 0.1 leaving 27 MSAs for the study (Fig. 1 & Table S1).

3.2. Sampling of NEI data

A metropolitan area consists of one or more counties that contain an urban core with a population $\geq 50,000$ (Census Bureau, 2015). Because the NEI data is at the county level, we used the geographical boundaries

of the 27 MSAs according to 2015 delineations for core based statistical areas (Census Bureau, 2015) to extract total population from the county level US Census population estimates, and total and highway CO emissions from NEI. In addition, we calculated per capita highway CO emissions for the 27 MSAs to study how per capita CO emissions changed over time.

3.3. Theil-Sen trend analysis

We used Theil-Sen trend analysis to detect significant trends in the quality filtered CO time-series using AK threshold of 0.1. The Theil-Sen trend analysis is robust against missing data and outliers (Helsel and Hirsch, 1995). For a set of n observations in a time series, Theil-Sen trend first computes n(n-1)/2 slope estimates $(y_j - y_i)/(t_j - t_i)$ for all $i < j$ and then take the median of all the estimates as the slope estimate of the trend, where y_j and y_i are the measurements at time t_j and t_i respectively (Sen, 1968; Theil, 1950). All statistical analyses were conducted in R (Version 3.2.2) and we used Version 1.6.5 'openair' package (Carslaw & Ropkins, 2012) for the Theil-Sen trend analysis.

4. Results

Both MOPITT surface CO and the NEI CO data exhibited significant, sublinear power-law scaling relationship with ambient population. The 15-year of MOPITT data across the 27 MSAs resulted in a log–log relationship between annual average CO concentrations and population with a coefficient of determination (r^2) of 0.30 and scaling coefficient (β) of 0.18 ($p < 0.001$) (Fig. 2 & Table 1). We also examined the scaling relationships for individual years. Scaling coefficients ranged from 0.15 to 0.20, with a mean of 0.18, and r^2 ranged from 0.32 to 0.46. The coefficients of determination gradually decreased from 2000 to 2015, with the highest r^2 of 0.46 for year 2000 and the lowest r^2 of 0.32 for 2015.

NEI CO data yielded stronger scaling relations with population than MOPITT CO retrievals, with r^2 of 0.58 and β of 0.74 for highway vehicle CO emissions, and r^2 of 0.71 and β of 0.77 for total CO emissions using all the data across five years (Fig. 3 & Table 1). Scaling relationships for individual years were stronger with $r^2 > 0.81$. β changed slightly from year to year, with β ranging from 0.73 to 0.80 for highway vehicle CO
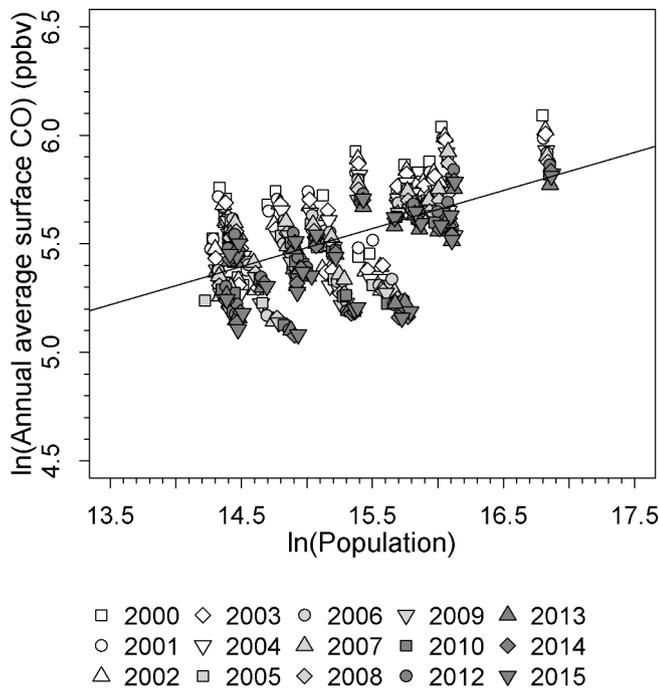
☐ 2000 ◇ 2003 ⊙ 2006 ▽ 2009 ▲ 2013
○ 2001 ▽ 2004 △ 2007 ◼ 2010 ◆ 2014
△ 2002 ◼ 2005 ◇ 2008 ⬤ 2012 ▽ 2015

Fig. 2. Log-log relationships between annual average surface CO concentrations and population using 15 years of data: ln(CO) = 0.18 × ln(population) + 2.9.

Table 1

Coefficients of determination (adjusted r²) and slopes (β) for the power-law scaling relationships between population and NEI total CO, NEI highway CO, and MOPITT surface CO.

	NEI total CO (ton)	NEI highway CO (ton)	MOPITT CO (ppbv)
r^2	0.71	0.58	0.30
β	0.77	0.74	0.18

emissions, and from 0.77 to 0.81 for total CO emissions. We found that β and intercepts were not significantly different from year to year.

Per capita highway CO emissions have been generally decreasing from 2002 to 2014 for the majority of the MSAs (Fig. 4). Several cities,

however, exhibit slightly different patterns in changes of per capita highway CO emissions. Per capita highway CO emissions in New Orleans increased in 2005, perhaps a consequence of Hurricane Katrina. Providence, Cincinnati, and Indianapolis experienced slight increases in per capita CO emissions in 2011. Rates of per capita CO emissions have slowed down since 2011 in New York City and Philadelphia. Chicago, Richmond, Virginia Beach, and St. Louis experienced slight increases in per capita CO emissions from 2011 to 2014.

Theil-Sen trend analysis on both MOPITT CO and the EPA ground measurements suggests significant negative CO trends over these cities. There is no significant correlation between Theil-Sen slope estimates from MOPITT CO and those from EPA CO data due to large differences in the scale between the two datasets (Fig. 5). Theil-Sen trend analysis on MOPITT CO showed that the largest CO reduction happened in New York City and Philadelphia, while *in situ* EPA CO data indicates that Hartford, CT, Nashville, TN, Raleigh, NC, and New York City experienced the greatest CO reduction among the 27 MSAs from 2000 to 2015 (Table S2).

5. Discussion

We found significant, sublinear scaling relationships between both CO concentrations and emissions with population. The sublinear relationships indicate larger MSAs emit less CO per capita than smaller MSAs. Highway vehicle CO emissions scaled similarly with total CO emissions, because highway vehicle CO emissions contributed most to total CO emissions in the US cities. It was reported that total traffic delays and transportation CO_2 scaled superlinearly with population due to congestion (Louf & Barthelemy, 2014). If there were more per capita traffic delays and higher per capita transportation CO_2 in larger cities than smaller ones, then we would expect to find per capita highway vehicle CO emissions also higher in larger cities. However, we find the opposite in terms of CO emissions and concentrations (Table 1). Traffic congestion, although an important factor, is one of several influences on emissions. The availability of public transportation, local air emission control policies, lower emission rates from newer vehicles may also contribute to emissions reduction in the transportation sector. For example, the continuing replacement of conventional internal combustion engines with hybrids, electric vehicles (EVs), and, eventually, hydrogen fuel cell vehicles should lead to lower CO and CO_2 emissions in cities. From the policy-makers side, governments have designed and implemented emission reduction programs to promote construction of alternative fuel fueling stations, and to encourage consumers to
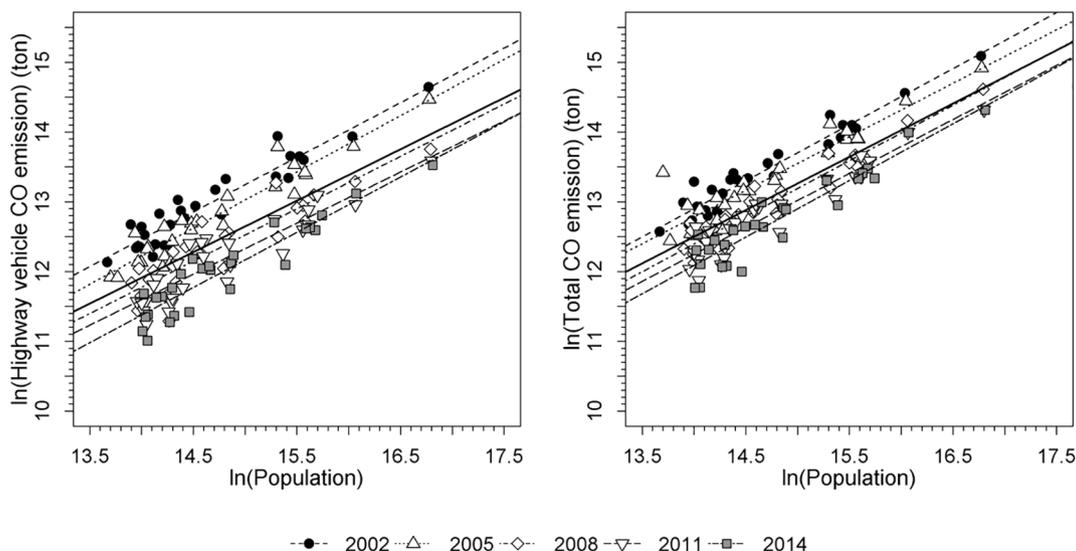

-•- 2002 ··△·· 2005 -◇- 2008 -▽- 2011 -◼- 2014

Fig. 3. Log-log relationships between CO emissions and population. Solid black line is the regression line for all observations across five years. Left figure for highway CO emissions: ln(CO) = 0.74 × ln(population) + 1.6; right figure for total CO emissions: ln(CO) = 0.77 × ln(population) + 1.7.
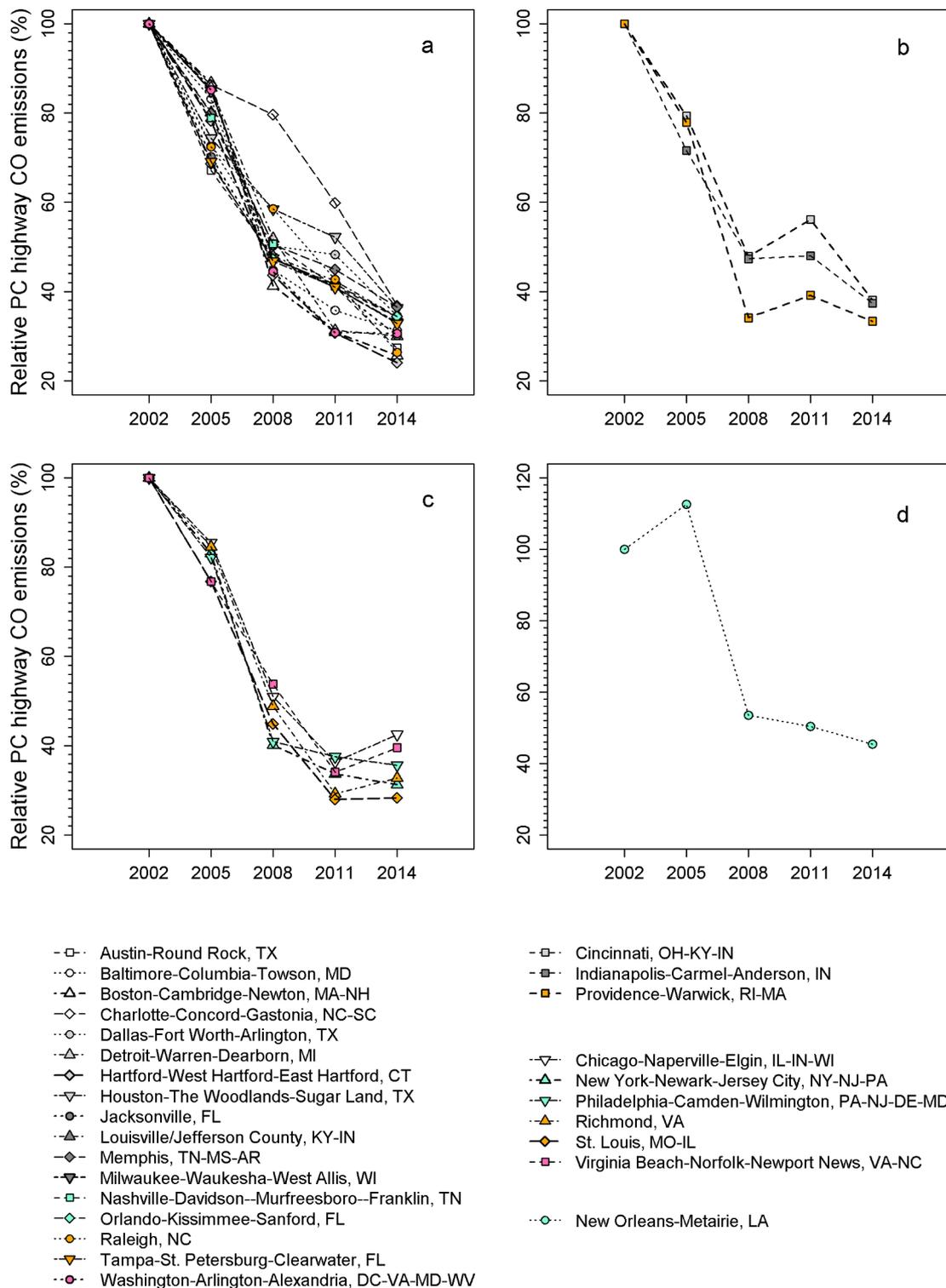
Fig. 4. Relative changes of per capita (PC) highway CO emissions that were normalized to the 2002 value for each city. Note different scaling for the y-axis in d.

purchase of EVs with government incentives or taxes exemption (Dia-mond 2009; Mabit & Fosgerau, 2011). At the state level, some programs are state-wide, while others target non-attainment areas that have air quality worse than the National Ambient Air Quality Standard (NAAQS) under the Federal Clean Air Act. For example, the Drayage Truck In-centive Program provides financial incentives for the replacement of older drayage trucks operating at seaports and Class I rail yards in non-attainment areas of Texas (TCEQ, 2018). Each state is responsible for developing a State Implementation Plan (SIP) to clean the air and meet

federal air quality standards (EPA, 2018a). The SIP addresses unique air pollution problems in each states. Among the 27 MSAs in this study, portion areas of 11 MSAs (Table S2) had been designated as CO non-attainment areas, but were all redesignated as attainment before April 2002 (EPA, 2018b). Each state, then, implements a maintenance plan, which is revised as the status of air quality changes, to continuously meet NAAQS. Thus, during our study period, eleven MSAs have a CO maintenance plan that targets a portion area of these MSAs, while the rest don't have a maintenance plan for CO, but may contain a
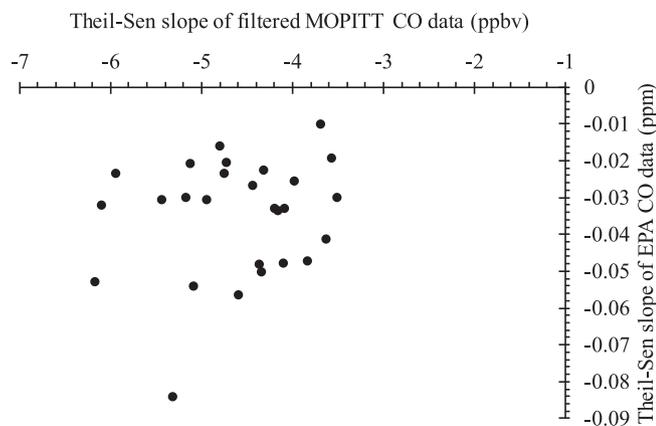
Fig. 5. A comparison of Theil-Sen slope estimates between filtered MOPITT CO and EPA CO data (Spearman r of 0.13 and p value of 0.52).

nonattainment/attainment area for other air pollutants. All CO nonattainment areas in the US have been redesignated to maintenance since September 27, 2010 (EPA, 2018c).

Larger cities tend to have a higher level of wealth where people may be more willing to invest in newer technologies that lead to greater reductions in air pollutants (Dodman, 2009). Slowik and Lutsey (2018) reported that San Jose area had the highest share of new plug-in EVs in 2017, and the share of new plug-in EVs in the top 50 US MSAs exceeds that of the rest of the US by a factor of about 2.5. Public transportation systems can be another important factor. For example, the extensive public transport system in New York City keeps the city's per capita emissions relatively low (Dodman, 2009). The New York-Northern New Jersey-Long Island, San Francisco-Oakland-Fremont, and Washington-Arlington-Alexandria are the top three metro areas that had the highest percentage of workers who commuted by public transportation in 2009 (McKenzie & Rapino, 2011). Large MSAs, such as San Francisco and Boston, experienced large declines in provide automobile commuting rates between 2006 and 2013 which can be ascribed to their better public transportation systems than other MSAs (McKenzie, 2015). Our findings imply that a MSA finds its way to produce less per capita waste as it becomes larger. From the urban planning side, compact and dense urban with sustainable transportation system seems to more favorable, as indicated by the significant negative correlations between per capita CO emission and population density (Fig. S2). Due to the limitation of the CO data, future research should use finer geospatial data to reveal the linkage between urban form, human activities, and their environmental impacts, to better support urban planners and designers. Nevertheless, urban planners and the transportation sector should continue their efforts in reducing traffic congestion and improving air quality, while electricity sector will benefit from innovative technology and clean energy sources (such as wind and solar) to meet increasing electricity demands and achieve sustainability (Soares et al., 2018).

Similar to stable scaling coefficients across time for CO_2 found by Fragkias et al. (2013), we found that the scaling coefficients for CO varied little on an annual basis, despite the relatively short CO lifetime in the troposphere (Novelli, Masarie, & Lang, 1998). Lamsal et al. (2013) found a β of 0.40 for the US cities between NEI NO_x and population. Our study yielded a β of 0.77 (0.74) between NEI total (highway) CO concentrations and population — a value that is close to three-quarters scaling relationship found between metabolic rate and body mass of a range of mammals (Kleiber, 1961). Similar to natural organisms, cities consume resources to maintain functioning and, thus, must excrete wastes that are a by product of the consumption process (Kennedy, Pincetl, & Bunje, 2011). Sublinear scaling relationships suggest that larger cities produce less per capita 'waste' in terms of CO and NO_2 emissions. A decline in per capita outputs of pollutants can be attributed to a combination of changes in public policy, improvements

in technology and public infrastructure, and impacts of increases in population density (Ngo & Pataki, 2008).

We found a weaker scaling relation from MOPITT surface CO than NEI CO data. This discrepancy is due, in part, to the methodological difference between observing concentrations and estimating emissions as well as to the coarse spatial resolution of MOPITT CO data and the detection limits of the MOPITT instrument. With an average atmospheric residence time of two months that is partially a function of insolation and, thus, latitude and time of year, it is more challenging to detect elevated CO over cities compared to shorter lived NO_2 detected by spaceborne sensors (Buchwitz, Khlystova, Bovensmann, & Burrows, 2007). Due to the constraints of the MOPITT surface CO data, our study used a relatively small number of MSAs to identify the scaling relationship between CO concentrations and population. CO vehicular emissions are well correlated with NO_x vehicular emissions, but the CO/NO_x emission ratio can vary over space and time (Parrish et al., 2002). Although our results are not directly comparable to those in Lamsal et al. (2013) who reported an r^2 of 0.50 for the scaling relation between OMI NO_2 and population for the US cities, it is not surprising that the scaling relation from MOPITT surface CO is weaker than that from OMI NO_2, given the coarser spatial resolution of MOPITT CO than OMI NO_2 data—1° by 1° vs. 0.25° by 0.25°, and increased difficulties in detecting enhanced levels of CO over MSAs compared to NO_2. Lamsal et al. (2013) found similar scaling relations from OMI NO_2 and the National Emissions Inventory data for NO_x. In contrast, we found a much smaller scaling coefficient for the MOPITT CO data than for NEI CO data. The significant scaling relations between satellite measurements of air pollutants (CO in this study and NO_2 from Lamsal et al., 2013) and population have demonstrated the possibility of using satellite observations of atmospheric trace gases to study urban scaling relationships. Oliveira et al. (2014) found higher scaling coefficients for CO_2 emissions using city boundaries defined by City Clustering Algorithm than using MSA boundaries and concluded that different definitions of city boundaries may result in different scaling coefficients. Therefore, different sampling methods on the MOPITT surface CO and NEI CO data may also in part attribute to the discrepancy between the scaling relations from the two datasets. Due to the coarse spatial resolution of the MOPITT data, we were unable to investigate the effects from the two different sampling approaches: 1° buffer for the MOPITT data and MSA boundaries for NEI. However, we can benefit from satellite observations with improved spatial resolution to evaluate the effect of city boundary on scaling relations.

Rybski et al., 2016 analyzed previous research and concluded that the scaling relation between CO_2 emissions and population size depends on economic development: super-linear (β > 1) scaling over cities in economically less developed countries, β = 1 over cities in economic emerging countries, and sub-linear (β < 1) scaling over cities in developed countries. Scaling coefficient (β) reflects the economic development of a country, and thus changes over time as a country's economic develops (Rybski et al., 2016). The sub-linear scaling between CO and population over the US MSAs is in line with previous research findings because the US is a developed country. With finer spatial resolution and better data quality of satellite measurements of air pollutants, it is possible to use a systematical and consistent approach to study how per capita air pollutant in cities changes as economic develops over many countries and over time. One possibility is to use Sentinel-5P satellite, a European Space Agency (ESA) satellite that was launched in October 2017 and provides 7 by 7 km spatial resolution and better data quality of air pollutants.

Trend analyses on both satellite and *in situ* CO data showed significant reductions in CO concentrations and emissions over the 27 US MSAs. Air quality, thus, has been improving over time in each individual city. As technology advances and becomes mature, more and more hybrids, electric vehicles, and hydrogen fuel cell vehicles will be entering city roadways. Reduction in CO and other air pollutants will continue. California and several other states have set a target to put 3.3

million electric and fuel cell vehicles on the road by 2025 (Hirsch, 2014). This multi-state zero-emission vehicle plan not only promotes reduction of CO, NO_x, ozone, and particulate matter in the air, but also aims to reduce greenhouse gas emissions. Sustainable urban planning and alternative transportation also play a significant role in reducing per capita air pollutants and greenhouse gas emissions. Government incentives, such as the "Cash for Clunkers" program boosted sales of newer, safer, cleaner, and more fuel-efficient vehicles, can speed up the 'greening' of the transportation sector (Blinder, 2008). All these efforts together lead to improved energy use efficiencies and reductions in per capita 'waste'. Per capita highway CO emissions, although in general decreasing over time for the 27 MSAs (Fig. 4), have experienced a slowdown in the rate of reduction since 2008, which may be attributable to the "Great Recession" that commenced in 2009. The slowdown in CO reduction may also be due, in part, to a technology plateau in reducing CO emissions from gasoline vehicles and replacement of older vehicles by zero- or near zero emission vehicles occurring at a slow pace.

6. Conclusion

This study investigated the scaling relationships between CO concentrations and emissions with ambient population over major US MSAs using MOPITT surface CO and NEI CO data. We found sublinear scaling relationships ($\beta < 1$) between CO emissions and ambient population, suggesting lower per capita CO emissions in larger than smaller MSAs. The two CO datasets did not exhibit similar scaling coefficients due in part to the difference in measurements between annual emissions in tons and annual average monthly surface concentrations in ppbv. The detection limits of the MOPITT instrument and coarse spatial resolution may explain why the scaling coefficient from MOPITT surface CO retrievals is smaller than NEI CO data. Finer spatial resolution and better data quality of CO satellite measurements are required to study its scaling relation with population. Future research can use the Sentinel-5P satellite data which provide better spatial resolution and data quality of air pollutants. Trend analysis of the long-term MOPITT surface CO retrievals revealed significant CO reduction from 2000 to 2015, and New York City, the largest city, experienced the strongest decreasing trend in CO (Table S2). The NEI CO data revealed that reduction in per capita CO emissions has been slowing down since 2008. In summary, CO emissions have been decreasing over time in each individual US MSA, and the larger MSAs appear more emissions efficient than smaller ones.

Acknowledgement

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.landurbplan.2018.12.009.

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