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Mobile Source CO_2 Mitigation through Smart Growth Development and Vehicle Fleet Hybridization

BRIAN STONE, JR.,*,†
ADAM C. MEDNICK,‡
TRACEY HOLLOWAY,§ AND
SCOTT N. SPAK‖

City and Regional Planning Program, Georgia Institute of Technology, Atlanta, Georgia, 30332-0155, Nelson Institute for Environmental Studies and Departments of Atmospheric and Oceanic Sciences and Civil and Environmental Engineering, University of Wisconsin–Madison, Madison, Wisconsin, Department of Urban and Regional Planning, University of Wisconsin–Madison, Madison, Wisconsin, and Atmospheric and Oceanic Sciences, University of Wisconsin–Madison, Madison, Wisconsin

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This paper presents the results of a study on the effectiveness of smart growth development patterns and vehicle fleet hybridization in reducing mobile source emissions of carbon dioxide (CO_2) across 11 major metropolitan regions of the Midwestern U.S. over a 50-year period. Through the integration of a vehicle travel activity modeling framework developed by researchers at the Oak Ridge National Laboratory with small area population projections, we model mobile source emissions of CO_2 associated with alternative land development and technology change scenarios between 2000 and 2050. Our findings suggest that under an aggressive smart growth scenario, growth in emissions expected to occur under a business as usual scenario is reduced by 34%, while the full dissemination of hybrid-electric vehicles throughout the light vehicle fleet is found to offset the expected growth in emissions by 97%. Our results further suggest that very high levels of urban densification could achieve reductions in 2050 CO_2 emissions equivalent to those attainable through the full dissemination of conventional hybrid-electric vehicle technologies.

Introduction

Cities in the United States are presently experiencing their most rapid rate of growth since before the Second World War (*1*). This growth is being experienced not only as a result of national population trends but in response to a shift in regional migration patterns away from exurban and suburban zones toward city centers. Projected U.S. population growth overall, coupled with the aging of the postwar Baby Boom, indicate metropolitan growth and urban reconcentration will increase in coming decades (*2*). To meet projected demand for housing and other building types, Nelson (*3*) predicts that approximately 50% of the built environment—streets, buildings, and parking lots—of U.S. cities in 2025 will have been newly developed or redeveloped since 2000. Such a massive redevelopment of urbanized regions over a relatively short period of time entails both challenges and opportunities. Accommodating an influx of new population growth promises to further strain limited air, energy, and water resources in metropolitan areas. However, extensive physical redevelopment of the built environment of urban centers presents an opportunity to measurably reduce per capita demand for these resources through more compact, mixed-use, and transit-supportive patterns of growth—the essence of so-called "smart growth."

This paper presents the results of a study on the effectiveness of smart growth development patterns and vehicle fleet hybridization in reducing mobile source emissions of carbon dioxide (CO_2) across 11 major metropolitan regions of the Midwestern U.S. over a 50-year period. During the past two decades, there has been considerable debate over the effectiveness of smart growth as a means of controlling gasoline consumption and vehicle emissions (see, for example refs *4*–*6*,). The development and dissemination of advanced vehicle technologies to improve fuel economy is widely viewed as the principal means of lowering greenhouse gas emissions from the transportation sector (*7, 8*). In a recent viewpoint in *Environmental Science & Technology*, however, Marshall (*9*) argues that urban design offers an "undervalued opportunity" for climate change mitigation. Here we evaluate this claim through the estimation of future mobile source CO_2 emissions associated with land development and vehicle fleet hybridization scenarios targeted to specific metropolitan regions.

Presently, hybrid-electric vehicles (HEVs) are the most fuel-efficient cars and light trucks available in the market, with miles per gallon (MPG) ratings ranging from 30 to 60% higher than comparable internal combustion-only vehicles (*10*). However, the lag-time for disseminating this and other advanced vehicle technologies throughout the U.S. fleet could be decades-long, due to high costs and slow rates of vehicle turnover and scrappage (*11*). Schafer and Jacoby (*12*) project that HEVs and other advanced vehicle technologies will continue to account for a small fraction of the U.S. vehicle fleet as late as 2030, even under an aggressive carbon tax. The most advanced vehicle technology in existence today, hydrogen fuel cell vehicles, could take 10–15 years to enter the commercial market (*13*).

Moreover, a number of empirical studies have shown that household vehicle miles traveled (VMT) increases in response to improvements in fuel economy—the so-called "rebound effect." Typical estimates of the long run (i.e., 10 years and thereafter) rebound effect are approximately 0.20 (*14*). This means that, historically, a 10% increase in average fuel economy corresponds to a 2% increase in average VMT. This rate appeared to decrease somewhat during the 1990s—perhaps reflecting the decreased sensitivity of VMT to changes in fuel economy in periods of relatively high real income and low fuel costs (*15*).

The relationship between varying land use development patterns and household daily travel behavior, including VMT, has been analyzed in a number of empirical studies (for extensive reviews, see refs *16*–*18*). Five dimensions of urban form, in particular, are believed to influence VMT and, by extension, vehicle emissions. These include: population density, land-use mix, and street-network design (*19*);

* Corresponding author phone: (404) 894-6488; fax (404) 894-1628; e-mail: stone@gatech.edu.
† Georgia Institute of Technology.
‡ Department of Urban and Regional Planning, University of Wisconsin–Madison.
§ Nelson Institute for Environmental Studies and Departments of Atmospheric and Oceanic Sciences and Civil and Environmental Engineering, University of Wisconsin–Madison.
‖ Atmospheric and Oceanic Sciences, University of Wisconsin–Madison.

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regional accessibility (*20*); and proximity to transit (*21*). The relationships between these variables and VMT are typically reported as elasticities; that is, the average percent change in VMT that can be expected to occur in response to a one percent increase, or decrease, in a given urban form measure.

VMT elasticities present a useful metric for gauging the impact of land use change, enabling the comparison of different variables for their relative influence on VMT. Ewing and Cervero (*17*) reported typical elasticities with respect to local density, land use mix, and design, of −0.05, −0.05, and −0.03, respectively, versus typical elasticities with respect to regional accessibility of −0.20. However, Stone et al. (*22*), employing an approach similar to that presented herein, estimated elasticities of −0.41 with respect to population density in "urban" tracts, versus −0.19 in "suburban" tracts. The variation in elasticities reported from different empirical studies points to the need for additional work in this field, including the development of improved data inputs, more consistent urban form measures, and improved model specifications (*16, 18*).

Methods

The methodology outlined below builds on an initial research paper (*22*), which considered a more limited number of land use scenarios and did not address the implications of vehicle technology change for mobile source emissions. In the present study, we set out to predict the relative and combined impacts of alternative urban development and vehicle technology futures on total mobile source emissions of CO_2. We associate future emissions with land development and vehicle fleet hybridization scenarios by integrating four different modeling components: (1) a set of tract-level demographic, socioeconomic, and land use projections based on alternative growth assumptions; (2) a household vehicle trip modeling framework, linking average household VMT to projected tract-level characteristics; (3) a CO_2 emissions estimation procedure developed by the U.S. Environmental Protection Agency (EPA); and (4) the development of a fleet-level emissions adjustment factor, based on the complete transformation of the light vehicle fleet from internal combustion-only to hybrid-electric vehicles by 2050. A brief description of these elements follows a description of the study region below.

Study Region. The study region is EPA Region V, which includes six Midwestern states: Illinois, Indiana, Michigan, Minnesota, Ohio, and Wisconsin. We report results for the eleven most populous metropolitan statistical areas (MSAs) in the region, as of 2000. These 11 areas vary considerably in their average tract-level density as of 2000, ranging from 2.4 persons per acre (Madison, Wisconsin) to 13.4 persons per acre (Chicago, Illinois), with a median density of 3.8 persons per acre. The region as a whole experienced slow overall population growth relative to other parts of the U.S (*23*). At the same time, it has experienced considerable urban deconcentration, particularly during the 1970s and early 1990s (*24*). We project that the populations of all 11 MSAs will increase over the next five decades, presenting the potential to significantly alter their geographic patterns of growth.

"Business-as-Usual" Development Scenario (BAU). The BAU scenario assumes future population and land use trends throughout the study region will be consistent with historical change. To derive this scenario, we extrapolated county populations to 2050 using a series of time-series regression models based on decennial census counts and annual intercensal estimates for the years 1970 through 2000 (*25*−*27*), and postcensal estimates for the years 2001 through 2004 (*28*). For each county we selected the projection with the lowest mean absolute percent deviation from the historic trend, extended to include independent projections for the years 2005 through 2030 (*29*). We then allocated future county population to tracts via constant-share apportionment, holding the 2000 county-to-tract population ratio constant. Each census tract was assigned a neighborhood type: "urban", "suburban", or "rural", based on a combination of its projected contextual density (i.e., its focal density, measured in persons per square mile in the individual tract and its surrounding tracts) and the projected contextual density of the tract's nearest regional center. Developed by the market research firm Claritas, Inc. (*30*), this approach is superior to the standard, dichotomous density thresholds provided by the U.S. Census Bureau for the purpose of predicting tract-level household travel (*31*). We assume tract-level employment and household income will follow historic and projected future trends and use regression-based methods similar to those used for population to extrapolate these variables to 2050. We further assume that household vehicle ownership will track changes in neighborhood type. A description of the process used to estimate vehicle ownership can be found in the Supporting Information (SI).

"Smart-Growth" Development Scenario 1 (SG1). In this scenario we assume the total population of each MSA in 2050 is the same as under BAU, but that new population added in each decade is reallocated away from rural to suburban and urban census tracts in response to the widespread adoption of growth management policies, such as urban growth boundaries (UGBs) and transit-oriented development. Specifically, we adjusted the proportion of projected BAU population growth occurring in each of the three neighborhood types to match historical development trends in Portland, Oregon, a metropolitan area with a widely recognized comprehensive growth management program. Between the 1980 Census (one year after the establishment of an UGB) and the 2000 Census, the proportion of Portland's population residing in urban and suburban census tracts increased at a significantly higher rate than Midwestern MSAs, on average, while the proportion of the population residing in rural census tracts increased by a significantly lower rate. Although the relative contribution of Portland's growth management policies to its increased urbanization is difficult to separate from the effects of other attributes, Portland nevertheless provides an empirical basis for modeling the effects of compact urban growth on emissions. Projected income and employment are assumed to be the same as under BAU, reflecting historic and projected subregional trends. Also, as with the BAU scenario, household vehicle ownership is linked to changes in neighborhood type.

"Smart-Growth" Development Scenario 2 (SG2). Under the SG2 scenario, a larger percentage of new MSA population growth is reallocated from rural to suburban and urban census tracts. Starting in the base year of 2000, all new population growth per MSA is directed to urban and suburban tracts only, with 10% of this growth directed to urban tracts and the remaining 90% directed to suburban census tracts in 2010. The urban allocation then increases by 10 percentage points per decade, rising to 50% of new population growth by 2050. For those MSAs without urban tracts as of 2000, 100% of projected population growth is apportioned to suburban tracts, until one or more tracts becomes urban, after which the process of incrementally increasing the urban growth share begins. Relative to the Portland-based SG1 scenario, SG2 results in higher urban populations and greater mean densities per census tract, as well as a larger number of tracts with densities of 10,000 people per square mile and higher. Income and employment values were assumed, once again, to be the same as BAU, while household vehicle ownership is again linked to changes in neighborhood type over time.

Vehicle Activity Modeling. The core of our approach is a vehicle activity modeling framework for associating future land use and demographic characteristics, under our BAU, SG1, and SG2 scenarios, with household vehicle travel. We extend a "transferability" framework, developed by researchers at the Oak Ridge National Laboratory (ORNL) to support the derivation of tract-level travel statistics from the 1995 Nationwide Personal Transportation Survey (NPTS) (*31*). This framework enables the estimation of VMT in response to three census variables: median household income, vehicle ownership, and employment rate, plus the neighborhood type classification discussed above. These variables are used to identify clusters of census tracts hypothesized to share similar travel characteristics. Once tracts are grouped into these clusters, average daily VMT per household is derived from NPTS responses in each cluster (SI Table S2) and used to estimate tract-level VMT based on the number of households per tract. For this study, we added an additional "super urban" cluster, comprising all census tracts with a contextual population density of 10,000 or more people per square mile, to capture high densities in future years.

We assume that as the demographic, socioeconomic, and land use characteristics of a tract change over time, its cluster designation—and thus its average household VMT—will change correspondingly, a process we refer to as "cluster-migration" (SI Figure S2). This framework has been shown to predict vehicle travel quite accurately. A comparison of VMT estimated using the framework to data obtained through independent travel surveys conducted in New York, Massachusetts, and Oklahoma found that the framework estimated VMT at the metropolitan and state levels with a mean error rate of approximately 3.1% (*31*).

Vehicle Emissions Modeling. The U.S. EPA's MOBILE6 emissions factor model provides the most widely used tool to estimate mobile source emissions of regulated pollutants (e.g., carbon monoxide, nitrogen oxides, and volatile organic compounds) in response to a set of vehicle fleet, operational, and climate characteristics. While MOBILE6 may be used to estimate CO_2 emissions, the model does not adjust CO_2 emission factors for variation in vehicle speed (*32*). Since average vehicle speed impacts fuel economy, and varies considerably among urban, suburban, and rural tracts, we chose to estimate CO_2 emissions independently, using a standard method published by the U.S. EPA (*33*). In this method, the typical passenger vehicle is assumed to emit 8877 g of CO_2 for every gallon of gasoline consumed. Tract-level CO_2 emissions were estimated as follows:

$$CO_2 = (\text{total VMT}/\text{average MPG}) \times 8.877 \text{ kg} \quad (1)$$

Average MPG values for urban, suburban, and rural census tracts, respectively, were derived from an ORNL study of a fleet-representative sample of cars and light trucks tested at varying speeds (*34*). Using eqn 1, we calculated tract-level CO_2 emissions from our alternative BAU, SG1, and SG2 VMT projections, and MPG values specific to neighborhood type (SI Table S3). We assume that the composition of the vehicle fleet is constant across the six state study region.

Hybrid-Electric Vehicle Fleet Scenario (HEV). In this scenario we assume that 100% of the study region's vehicle fleet will be HEVs by 2050—half of which will be grid plug-in HEVs—but that the fleet will otherwise remain unchanged from 2000 in terms of its relative composition of different vehicle size classes (*36*). As noted above, there are several challenges to achieving widespread market penetration of HEVs. A number of investigators have predicted only modest success in the midterm (e.g., refs *11, 12*). Our principal objective here, however, is to compare and evaluate the sensitivity of CO_2 emissions to smart growth, in relation to an advanced technology scenario, under the most optimistic assumptions.

For our BAU scenario, we assume stock fleet fuel economy will increase from 19.5 MPG in 2000 to 25.6 MPG in 2050, which we projected via a time-series regression of the thirty-year "reference" forecast of U.S. stock fleet fuel economy developed by the U.S. Energy Information Agency (*35*). We then derived and applied an HEV emissions adjustment factor based on a study of the potential impacts of alternative technology futures on energy consumption over a 50-year time horizon (*36*). This study predicted that a complete hybridization of the U.S. light vehicle fleet by 2050 would result in an average stock fleet fuel economy of 33.0 MPG—representing a 29% increase over the conventional 2050 vehicle fleet. We assume that households will respond in turn by increasing their average daily VMT by 5.8% relative to the base-case, reflecting the historic rebound effect of 2% for every 10% increase in MPG (*15*). Based on these assumptions, we calculated our HEV emissions adjustment factor as follows:

$$\text{Emissions adjustment factor} = (MPG_{base}/MPG_{HEV}) \times (1 + \text{VMT Rebound}) \quad (2)$$

Results

Population Density. The extrapolation of historical growth patterns between 2000 and 2050 through the BAU scenario was found to increase the average metropolitan population density by almost 8% for the median city. This finding suggests that some degree of urban compaction, and therefore reduced rates of household VMT, would be likely to occur absent any policy intervention designed to increase population densities above historical trends.

The application of historic growth shares from Portland to Midwestern cities (SG1) resulted in an approximate 36% increase in the median metropolitan population density relative to the base year of 2000, a rate of increase more than four times greater than that of BAU. An increase in the urban growth share of 10 percentage points per decade (SG2) was found to produce an increase in the median population density of about 61% over the base year level, or a rate of increase almost seven times historical rates.

Vehicle Miles of Travel. The effects of each urban development scenario on VMT by region are presented in Figure 1. As the population density of an individual census tract increases over time it may "migrate" from a higher to a lower vehicle travel cluster, resulting in a lower rate of household VMT. For the median MSA of Grand Rapids, MI, total VMT increased by 64% between 2000 and 2050 under the BAU scenario. The SG1 and SG2 scenarios were found to increase median VMT by 56% and 47%, respectively. The more aggressive of the two smart growth scenarios, therefore, would result in a rate of increase in VMT about 26% lower than BAU.

At the individual MSA level, the percent increase in VMT over base year levels ranges from a low of about 14% in Dayton, OH to more than 100% in Madison, WI. The wide variability in VMT change by region is principally attributable to variation in population growth rates. While Dayton is expected to grow by less than 1.2% between 2000 and 2050, Madison's population is projected to almost double in size, resulting in a much larger increase in total vehicle travel.

CO_2 Emissions. The median change in VMT and CO_2 emissions by growth scenario are reported in Figure 2. Overall, the rate of change in CO_2 emissions between 2000 and 2050 is much lower than that of VMT across each of the three scenarios. As discussed above, future year estimates of CO_2 emissions are responsive to both changes in the regional size and distribution of population, as well as to expected improvements in the fuel economy of the conventional vehicle fleet. An increase in the vehicle fleet average fuel economy from 19.5 to 25.6 MPG serves to lower the rate of increase in CO_2 emissions relative to changes in total vehicle travel. As indicated in Figure 2, the BAU scenario produces

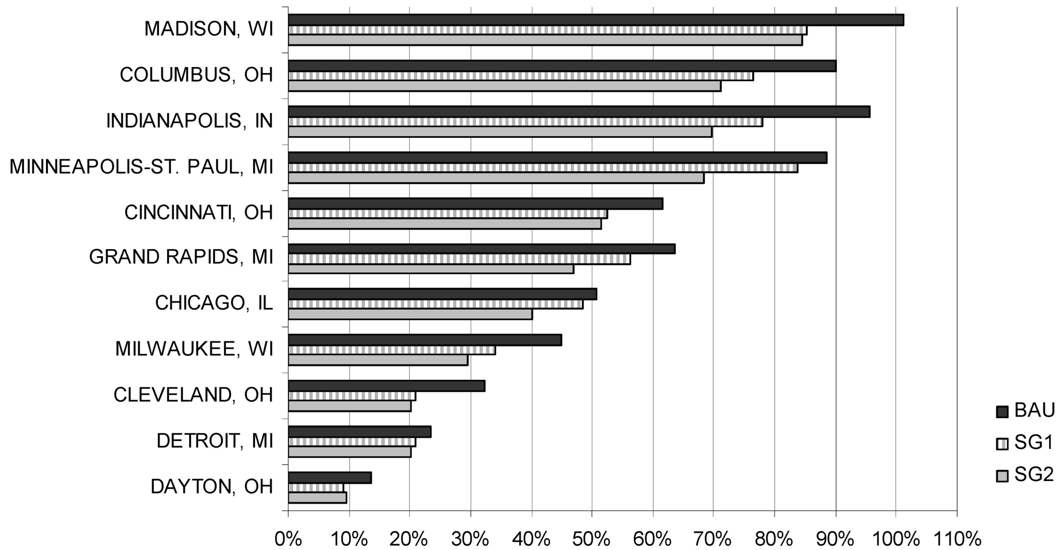

FIGURE 1. Percent change in VMT by MSA and scenario relative to 2000.

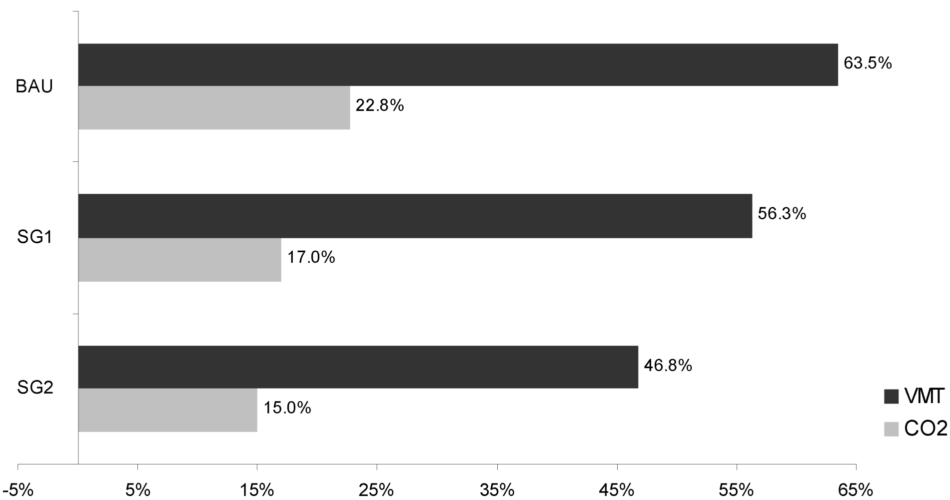

FIGURE 2. Percent change in median VMT and CO_2 relative to 2000.

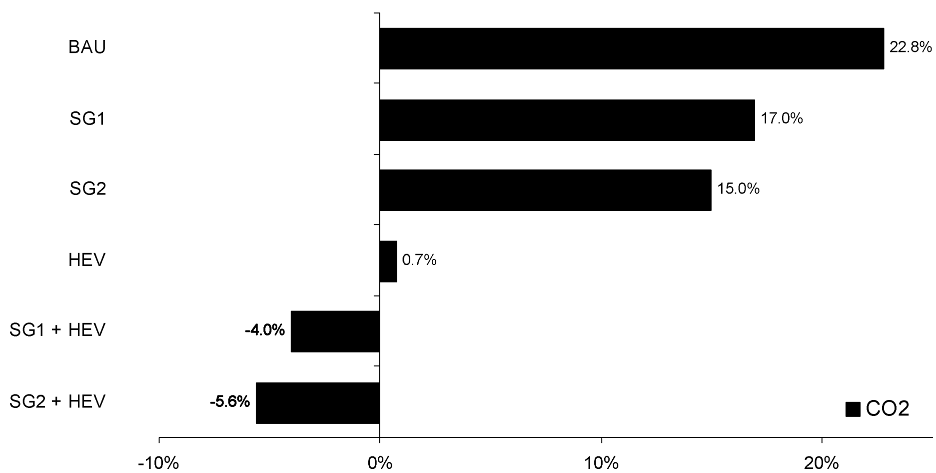

FIGURE 3. Percent change in median CO_2 by scenario relative to 2000.

an increase in emissions over base year levels approximately six percentage points higher than SG1, and approximately eight percentage points higher than SG2. The SG2 scenario results in a 34% reduction in the rate of growth in CO_2 expected under BAU.

Full hybridization of the light duty vehicle fleet by 2050 was found to be a more effective stand alone strategy for offsetting future CO_2 emissions than either of the two modeled smart growth scenarios. Figure 3 reports the same CO_2 trends for the BAU and smart growth scenarios presented in Figure 2, alongside the median results for the fleet hybridization and combined land use and technology change scenarios. The fleet hybridization scenario was associated with a 97% reduction in the expected growth in CO_2 emissions under the BAU scenario, effectively offsetting all of the impacts of future population growth on carbon emissions. Under the

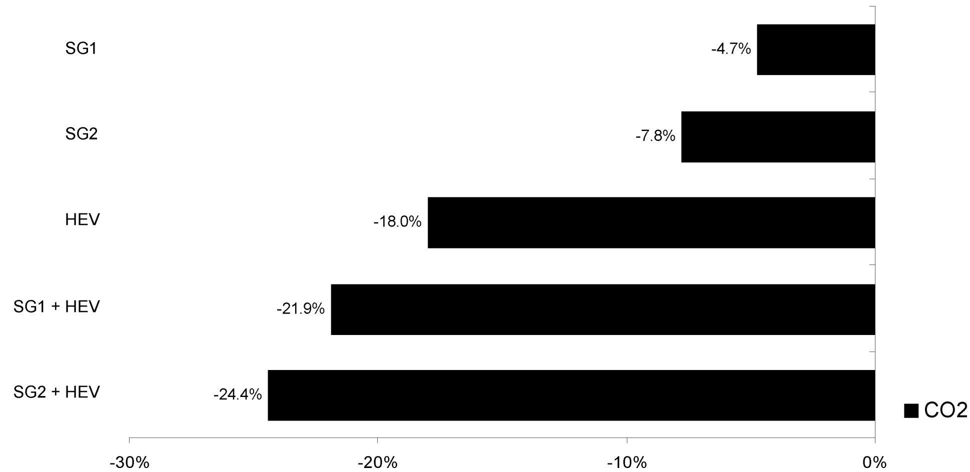

FIGURE 4. Percent change in median CO_2 by scenario relative to BAU.

combined SG1 and HEV scenario, CO_2 emissions are 4% lower than 2000 levels, and emissions resulting from a combined fleet hybridization and SG2 scenario are about 6% below base year levels.

Because improvements in vehicle fuel economy are reflected through both the smart growth and HEV scenarios, it is important to quantify the influence of land use change independent of technological improvements in the conventional vehicle fleet. Figure 4 presents changes in CO_2 emissions relative to the 2050 BAU scenario, rather than with reference to the base year of 2000. In doing so, fuel economy improvements in the conventional vehicle fleet and growth in population over time are held constant across the various scenarios, permitting the effects of land use change to be isolated and compared directly to additional fuel savings achieved through the HEV scenario. In this analysis, the HEV adjustment factor reflects an improvement in fuel economy over the 2050 conventional fleet average of 25.6 MPG, rather than over the 2000 conventional fleet average of 19.5 MPG.

With respect to the 2050 BAU scenario, the HEV scenario is found to be roughly twice as effective as the SG2 scenario in reducing emissions of CO_2. The Portland-like growth patterns reflected in the SG1 scenario are found to reduce BAU emissions in the median city by about 5%, while the more aggressive SG2 scenario reduces BAU emissions by about 8%. Full hybridization alone produces a reduction in BAU emissions of 18%. In combination, fleet hybridization and aggressive smart growth produce an almost 25% reduction in BAU emissions of CO_2.

Vehicle Travel and Emissions Elasticities. In concert, the rate of change in population density and vehicle travel may be used to compute VMT and emissions elasticities by MSA. The derivation of these measures is particularly useful in that it provides a means of estimating the likely change in VMT and emissions in response to any projected change in population density—not solely those modeled through the three growth scenarios developed for this study.

The median VMT elasticity for both the SG1 and SG2 scenarios was found to be −0.34, indicating that each 10% increment in population density resulting from these scenarios was found to be associated with a 3.4% reduction in household VMT. The responsiveness of CO_2 to density change was found to be slightly lower than that of VMT, averaging −0.30 for the median MSA under the SG1 (−0.29) and SG2 (−0.31) scenarios. These findings suggest that, all else being equal, a doubling of population density would result in an approximate 30% reduction in household vehicle CO_2 emissions in the median city, independent of improvements in vehicle technologies. The close correspondence in the elasticity values by scenario indicates that the relationship between density change and vehicle travel is largely insensitive to the magnitude of density change, at least with respect to the range of density change captured in the two smart growth scenarios. If true, the degree to which urban compaction would reduce VMT and emissions is directly dependent upon the degree to which a metropolitan area can increase density over a stated period of time.

Discussion and Policy Implications

The results of our study suggest the potential for both smart growth and technology change to measurably offset the growth in mobile source CO_2 emissions projected to occur within large metropolitan areas by 2050. Across the 11 Midwestern cities in the study, the more aggressive of the two smart growth scenarios was found to reduce BAU emissions of CO_2 by 8%, at the median, while the full dissemination of hybrid electric vehicles was found to reduce BAU emissions of CO_2 by 18%. These results highlight a number of important implications for the development of a more integrated air quality and climate change management strategy in large U.S. cities.

Emissions Reductions through Smart Growth. Although the CO_2 emissions reductions found to accrue under the two smart growth scenarios were lower than that of fleet hybridization as a stand alone strategy, our findings suggest urban densification holds the potential to approach or even surpass the benefits of conventional hybrid technologies if region-wide densities can be increased beyond the levels modeled herein. Our results suggest that, all else being equal, a doubling of mean population density throughout the median metropolitan area would have the effect of reducing vehicle CO_2 emissions by about 30% relative to the BAU scenario, while the full dissemination of conventional hybrid technology was found to reduce vehicle CO_2 emissions by 18%.

How feasible is a doubling of population density across a large metropolitan area relative to the 2050 BAU level? For the metropolitan area found to have the median CO_2 elasticity—Grand Rapids, Michigan—average metropolitan population density increases significantly under the BAU scenario by 2050 (63%), due to relatively high rates of population growth. Base year population densities in Grand Rapids would need to increase by a factor of about 3.3 to achieve a doubling of 2050 BAU levels. To attain the same level of CO_2 reductions relative to BAU as full fleet hybridization, average population densities in Grand Rapids would need to increase by about 160% over 50 years—an increase from about 3 to 7.8 persons per acre. While this rate of densification would represent a substantial increase over historical trends, the resulting average densities would roughly approximate those found today in Minneapolis-St. Paul, MN.

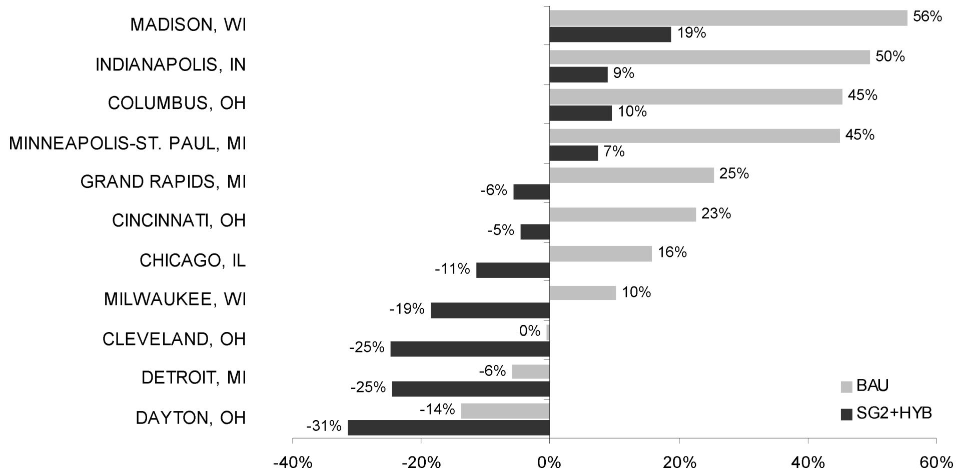

FIGURE 5. Percent change in CO_2 by MSA relative to 2000.

It is important to note that the historical growth rates employed in this study are reflective of a base period (1970−2000) for which the past decade was characterized by historically low energy prices relative to household income. Should energy prices increase rapidly in proportion to real household income, it is reasonable to expect a greater sensitivity of vehicle travel to land use change than that demonstrated through this study, as well as a more favorable political and economic environment for compact growth. With recent observations in mind, significant increments in metropolitan population density levels by 2050 may be feasible.

Emissions Reductions through Vehicle Fleet Hybridization. The results of our analysis suggest that continued dissemination of conventionally available hybrid-electric engine technology would significantly offset the growth in vehicle emissions above base year levels independent of land use change. Should the full hybridization of the light duty vehicle fleet take place by 2050, it would offset virtually all of the expected growth in CO_2 emissions under the BAU scenario.

An important contrast between the technological and smart growth strategies considered, however, is the implications of each for a wider range of problems associated with urban growth and vehicle use, such as traffic congestion, disinvestment in transit, and low rates of physical activity (*37*), as well as the life-cycle energy and pollution associated with the manufacture and disposal of vehicles (*38*). In light of the well documented problems associated with vehicle ownership and use in cities, climate change management strategies employing approaches rooted in both land use and technology change hold the potential to achieve the greatest offsets in mobile source emissions while addressing a wider array of problems associated with the growth of the vehicle population.

As illustrated in Figure 5, a climate change management strategy integrating the SG2 and HEV scenarios was found not only to significantly offset the growth in CO_2 emissions projected under BAU but to decrease these emissions beyond base year levels in 7 of the 11 metropolitan areas included in the study. This finding highlights the variable benefits of land use and technology change across economically diverse metropolitan areas. In those regions experiencing lower rates of population growth, relatively small offsets in new emissions hold the potential to measurably reduce regional carbon footprints. In those regions experiencing rapid rates of population growth, even substantial offsets in future vehicle travel and emissions may still result in an expansion of regional carbon footprints.

To date, our findings support the development of air quality and climate change management programs designed to both continue the dissemination of advanced vehicle technologies and promote smart growth urban development patterns to concentrate new population growth in urban centers. In contrast with a handful of previous studies (*6, 39, 40*), this research finds vehicle travel and emissions to have a reasonably high sensitivity to land use change over time and that such an approach presents a viable complement to strategies designed to reduce vehicle emissions through technological improvements in engine performance. In light of these findings, we recommend that large metropolitan regions of the U.S. pursue both sets of strategies through long-range land use, transportation, and air quality planning.

Acknowledgments

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Supporting Information Available

A description of the methods used for estimating and validating household vehicle ownership, along with expanded discussion on neighborhood type, urban form, and VMT, and assumed fleet characteristics; two figures; three tables. This material is available free of charge via the Internet at http://pubs.acs.org.

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