High Resolution Fossil Fuel Combustion CO$_2$ Emission Fluxes for the United States

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Quantification of fossil fuel CO$_2$ emissions at fine space and time resolution is emerging as a critical need in carbon cycle and climate change research. As atmospheric CO$_2$ measurements expand with the advent of a dedicated remote sensing platform and denser in situ measurements, the ability to close the carbon budget at spatial scales of ∼100 km^2 and daily time scales requires fossil fuel CO$_2$ inventories at commensurate resolution. Additionally, the growing interest in U.S. climate change policy measures are best served by emissions that are tied to the driving processes in space and time. Here we introduce a high resolution data product (the "Vulcan" inventory: www.purdue.edu/eas/carbon/vulcan/) that has quantified fossil fuel CO$_2$ emissions for the contiguous U.S. at spatial scales less than 100 km^2 and temporal scales as small as hours. This data product, completed for the year 2002, includes detail on combustion technology and 48 fuel types through all sectors of the U.S. economy. The Vulcan inventory is built from the decades of local/regional air pollution monitoring and complements these data with census, traffic, and digital road data sets. The Vulcan inventory shows excellent agreement with national-level Department of Energy inventories, despite the different approach taken by the DOE to quantify U.S. fossil fuel CO$_2$ emissions. Comparison to the global 1° × 1° fossil fuel CO$_2$ inventory, used widely by the carbon cycle and climate change community prior to the construction of the Vulcan inventory, highlights the space/time biases inherent in the population-based approach.

Introduction

Improving the quantitative understanding of the global carbon cycle has emerged as a central element in advancing our understanding of climate change and climate change projections, not to mention deepening our understanding of ecosystem level biogeochemical principles (*1*). Recent research has highlighted the importance of feedbacks between climate change and carbon uptake in the oceans and land, emphasizing the considerable spread in projected atmospheric CO$_2$ concentration due to uncertainties in surface−atmosphere exchange (*2*). The single largest net flux of carbon between the surface and the atmosphere is that due to the combustion of fossil fuels and cement production, recently estimated at 8.4 PgC year^{-1} (U.S. share is 1.6 PgC year^{-1}) for the year 2006 (*10, 3*). More importantly, quantitative assessment of biotic exchange on land and exchange with the oceans relies critically on the accuracy of both the incremental change of CO$_2$ in the Earth's atmosphere and the fossil fuel carbon flux from the surface. This is due to the fact that the surface−atmosphere exchange, particularly that between the terrestrial biosphere and the atmosphere, is commonly solved as the residual in large-scale budget assessments, such as atmospheric inversions (*4*).

Fossil fuel CO$_2$ inventories began as an accounting exercise based on the production/consumption of fossil fuels at the national scale (*5*). In most cases, little subnational allocation of the emissions was performed because the initial purpose, understanding 20th century global climate change, required little subnational information. Thus, the most common spatiotemporal distribution of fossil fuel CO$_2$ emissions occurred at an annual time scale and at the national spatial scale. Starting in the 1980s, research was begun to further subdivide these emissions into finer spatial and temporal scales (*6*). By the beginning of the 21st century, fossil fuel CO$_2$ emissions had been produced which were resolved globally, at the 1° × 1° spatial scale and most commonly at an annual time scale (*7, 8*). This subnational downscaling in space, however, was achieved through a spatial proxy such as population density statistics. The most recent work, prior to the results reported here, has quantified emissions at the scale of U.S. states/monthly (*9−13*) with two studies estimating and analyzing CO$_2$ fluxes from the power production sector down to the facility level (*14, 15*).

In the past decade, there has been a growing need, from both the science and policymaking communities, for quantification of the complete fossil fuel CO$_2$ emissions at space and time scales finer than what has been produced thus far (*16, 17*). Carbon cycle science requires more accurate and more finely resolved quantification because of downscaling of carbon budget and inverse approaches, which use space/time patterns of atmospheric CO$_2$ to infer exchange of carbon with the oceans and the terrestrial biosphere (*18*). These scientific needs have contributed to the launch of the Greenhouse Gases Observing Satellite (GOSAT), launched in early 2009 which will soon return measurements of the column concentration of atmospheric CO$_2$ with a instantaneous field of view of roughly 10 km and a 3 day return time (www.jaxa.jp/projects/sat/gosat/. The policymaking community in the U.S. has recognized the need for accurate, highly resolved CO$_2$ emissions due to the emerging requirements of proposed carbon trading systems or sectoral emissions caps (*19*). To answer this growing need for better resolution, accuracy, and linkage to the underlying emission drivers, research was begun on the Vulcan project. This paper serves as the first complete description of the methods and results emerging from this effort in which U.S., process-driven, fuel-specific, fossil fuel CO$_2$ emissions were quantified at scales finer than 100 km^2/hourly for the year 2002. We present the data sources and methods used to quantify fossil fuel CO$_2$ and the techniques used to perform spatial and temporal allocation. We quantify the results across a number of different data set dimensions and compare these results to inventories built at coarser scales. Lastly, we describe the implications for carbon cycle science by quantifying the

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Data Source	National Emissions Inventory				ETS/CEM	NMIM NCD	Aero 2k
Data Type	Non-road	Non-point	Point	Airport	Power Prod.	On road	Aircraft
Pollutant Utilized	Activity/population	CO	CO	CO	CO_2	VMT/population	CO_2
Incoming Resolution: Space/Time	County; Monthly	County; Annual	Lat/Lon; Annual	Lat/Lon; Annual/summer	Lat/Lon; Hourly	County; Monthly	$1°X 1°$; Monthly
Condition Data		Census; EIA sector/state/fuel/month sales	EIA sector/state/fuel/month sales			GIS Road Atlas; Mobile 6.2	
Final Resolution: Space/Time	County; Monthly	Census tract; Month	Lat/Lon; Month	Lat/Lon; Annual	Lat/Lon; Hourly	Road segment; Hourly	$1°X 1°$; Monthly
Sector	Mobile	Comm. Res. Ind. Electric	Comm. Res. Ind. Electric	Mobile	Electric	Mobile	Mobile

CO_2 Emissions

FIGURE 1. Data sources, incoming/outgoing resolution, conditioning data sets, and processing details used in the Vulcan inventory construction.

differences between the Vulcan inventory and the widely used predecessor inventory in terms of atmospheric CO_2 concentrations.

Methods

Data Sources. The Vulcan U.S. fossil fuel CO_2 emissions inventory is constructed from seven primary data sets with additional data used to shape the space/time distribution. A schematic is shown in Figure 1. A detailed description of the data sources, processing, and space/time allocation are provided in Supporting Information [SI Text 1]. The following is a summary of the Vulcan inventory methods.

The point, nonpoint, and airport data files come from the Environmental Protection Agency's (EPA) National Emissions Inventory (NEI) for the year 2002 which is a comprehensive inventory of all criteria air pollutants (CAPs) and hazardous air pollutants (HAPs) across the United States (24).

Point sources are stationary emitting sources identified to a geocoded location and comprise entities such as industrial facilities in which emissions exit through a stack or identifiable exhaust feature (25, 26). The area or nonpoint source emissions (dominated by residential and commercial activity) are stationary sources that are not inventoried at the facility level and represent diffuse emissions within an individual U.S. county reported as annual totals. The airport source includes emissions associated with geocoded airport locations (3865 facilities) and represent takeoff/landing cycle, taxiing, idling, and related aircraft activities on an annual basis though some airports report emissions only during summer months (27). In all three of these categories, we utilize the reported CO emissions.

Emissions due to aircraft, beyond the takeoff/landing cycle and emissions captured in the NEI airport database, are taken directly from the Aero2K aircraft CO_2 emissions inventory, defined on a global three-dimensional $1° \times 1°$ degree grid (28).

Because of the reliability of direct CO_2 monitoring and the need for fine time resolution data, we utilize CO_2 emissions available at electrical generating units (EGUs) reported to the EPA's Clean Air Market Division (CAMD) (United States Environmental Protection Agency, Clean Air Markets - Data and Maps, http://camddataandmaps.epa.gov/ gdm/index.cfm, accessed June 10, 2008) under Part 75 of the Clean Air Act (39). This data contains a large number of facilities that utilize Emission Tracking System/Continuous Emissions Monitoring systems (ETS/CEMs), widely considered the most accurate for CO_2 emissions estimation at these facilities (15).

The onroad mobile emissions are based on a combination of county-level data and standard internal combustion engine stochiometry. The county-level data comes from the National Mobile Inventory Model (NMIM) County Database (NCD) for 2002 which quantifies the vehicle miles traveled in a county by month, specific to vehicle class and road type (29). The Mobile6.2 combustion emissions model is used to generate CO_2 emission factors on a per mile basis given inputs such as fleet information, temperature, fuel type, and vehicle speed (30, 31).

Nonroad emissions are structured similarly to the onroad mobile emissions data and consist of mobile sources that do not travel on designated roadways. These data, retrieved from the NMIM NCD, have a space/time resolution of county/ month and are reported as activity (number of hour/month vehicle runs), population, and a CO_2 emission factor specific to vehicle class (27, 32).

Emissions Calculation. For data sets that do not directly provide CO_2 emissions, CO and CO_2 emission factors are used. These factors are specific to the combustion process and the 48 fuels tracked in the Vulcan system. CO emission factors are often supplied in the incoming data sets but are often missing or inconsistent with independent data. In many cases, therefore, standard emission factor databases are used to assign values to each combustion technology/fuel combination (Technology Transfer Network Clearinghouse for Inventories & Emissions Factor, WebFIRE, December 2005, http://cfpub.epa.gov/oarweb/index.cfm?action=fire.main, accessed 06/10/08). (33, 24). Where standard factors are not available, default emission factors are used (see Supporting Information, SI text 1 and SI Tables 1 and 2).

Emission factors for CO_2 are based on the fuel carbon content and assume a gross calorific value or high heating value, as this is the convention most commonly used in the U.S. and Canada (35). The basic process by which CO_2 emissions are created is as follows:

$$C_f^P = \frac{12}{44}\frac{PE_f^P}{PF_f^P}CF_f^P \qquad (1)$$

where C is the emitted amount of carbon, PE is the equivalent amount of uncontrolled criteria pollutant emissions (CO emissions), p is the combustion process (e.g., industrial 10 MMBTU boiler, industrial gasoline reciprocating turbine), f is the fuel type (e.g., natural gas or bituminous coal), PF is the emission factor associated with the criteria pollutant, and CF is the emission factor associated with CO_2. Percent oxidation level is embedded in the CO_2 emission factor.

Spatial/Temporal Downscaling. For those data sources that are not geocoded (mobile and nonpoint sources), allocation of emissions in space is performed through the use of additional data sets. Downscaling of the residential and commercial emissions in addition to the small amount of industrial sector emissions reported in the nonpoint NEI files are performed through use of U.S. Census tract-level (a U.S. Census tract is a geographic unit, smaller or equal to a U.S. county, defined for the purpose of taking a census) spatial surrogates prepared by the EPA (36).

Onroad emissions are also spatially downscaled from the county level by allocating the hourly/county/road/vehicle-specific CO_2 emissions onto roadways using a GIS road atlas (NTAD 2003 a collection of spatial data for use in GIS-based applications, United States. Bureau of Transportation Statistics, www.worldcat.org/oclc/52933703&referer=one_hit). The monthly/county/road/vehicle-specific CO_2 emissions are further subdivided in time using weight in motion (WIM) data obtained from the San Jose Valley traffic department (37).

Temporal downscaling to monthly time increments increments by state for the residential and commercial sector was performed for the nonpoint data sources by state and sector. Data on state-level, sector-based natural gas delivered to consumers from the DOE/EIA (38) was used to construct monthly fractions for the year 2002.

In order to facilitate atmospheric transport modeling, all of these emission sources are placed onto a common 10 km × 10 km grid. Geocoded sources are evenly spread across the resident grid cell while onroad sources are broken at the edges of the grid cells and summed into the cells to which they belong. Nonpoint sources are downscaled from the Census tract to 10 km via area-based weighting.

Results and Discussion

Figure 2 shows the Vulcan total 2002 U.S. fossil fuel CO_2 emissions, represented on a 10 km × 10 km grid. Emissions are most readily associated with population centers, but interstate highways and concentrations of industrial activity are also evident. Because the incoming data represents a mixture of spatial resolution, the gridded 10 km emissions map has both a background palette of uniform county-level emissions and higher resolution points and line sources. This reflects data that was reported at geocoded points, data that was reported at the county level but downscaled to the census tract level, and data which could not be further downscaled from the county scale. This last category shows up as emissions evenly distributed over county areas. This is particularly noticeable in the Plains states and intermountain West, where county sizes are large, population is low, and fewer industrial and electric production point sources exist.

The total emissions can be separated into contributions from standard economic sectors. Figure 3 shows the fossil fuel CO_2 emissions for the industrial, electric production, transportation, and the combined residential/commercial sectors. As with Fhe total emissions, the spatial distribution of the sectoral emissions reflect the underlying spatial nature of the economic sector and the mix of resolutions available

in the data sources. The industrial emissions in Figure 3a are derived mostly from the point source data and hence represent a large proportion of geocoded locations. Some industrial emissions, however, are derived from nonpoint emission data sources and represent 31.4% of the total industrial fossil fuel CO_2 emissions in the U.S.. Much of this (25.8%) is reported from the Texas oil and gas production industry. This is evident in the spatial distribution of industrial emissions in the state of Texas where these nonpoint emissions are spread evenly across the reporting county. A similar visual artifact is evident in Utah, Figure 3b, where the state chose to report all industrial sources solely as geocoded points and hence no county-spread industrial sources are denoted.

Figure 3b shows the CO_2 emissions from the electricity production sector which represents all fossil fuel electricity producing facilities. The vast majority of these emissions are reported through the ETS/CEMs reporting (97.5%), but a small amount of independent and small power producers are reported through local air pollution regulations (2.5%).

The mobile emissions in Figure 3c include emissions due to onroad, nonroad, and aircraft sources. As described in the methods, onroad emission sources are allocated to county-level roadways based on the twelve available road classes. Nonroad sources were distributed evenly throughout the reporting county, and aircraft sources (landing/takeoff and airborne) are allocated to geocoded airport locations. Onroad emissions are the dominant source within the mobile sector, representing 79.4% of the total, followed by aircraft emissions (11.8%) and nonroad emissions (8.8%). Both the density of urban onroad emissions and the presence of interstate travel are evident in Figure 3c as is the importance of airport emissions.

Figure 3d represents the sum of residential and commercial emissions. Both emission sectors follow population density with the residential sector somewhat less concentrated in population centers than commercial emissions. This is likely due to the fact that commercial buildings and businesses are more tightly related to urban development and employee density whereas residential structures are more dispersed into the suburban and exurban landscape.

Figure 4 displays the national-level monthly emissions for each of the five economic sectors. The monthly pattern of the largest emitting sector, electricity production, exhibits a peak in the months of July and August dominated by air-conditioning demand (15). Mobile emissions also exhibit a summer peak, though of a weaker magnitude. Industrial emissions show the least seasonality while both commercial and residential emissions have a summer minimum associated with a reduction in space heating during summer months.

Comparison to National-Level Estimates. The Vulcan inventory can be compared to independent estimates at aggregated scales. For example, estimation of annual fossil fuel CO_2 emissions at the national spatial scale is performed by the Department of Energy's Energy Information Administration (DOE/EIA) (9). Furthermore, the DOE/EIA defines emissions by economic sector and fuel category. Table 1 shows a comparison between the national-level DOE/EIA and Vulcan estimates by economic sector. Because the DOE/EIA does not disaggregate the transportation emissions in the same subcategories as done in the Vulcan inventory, estimates from the EPA (10) are also included for this sector. The EPA utilizes much of the DOE/EIA data to construct their estimates, but small differences do exist.

The DOE/EIA estimates are based on fuel supply surveys completed by producers or suppliers of energy sources and are tracked by fuel category as opposed to being primarily tracked by combustion category, as is the case in the Vulcan process. DOE/EIA measures the quantities of energy pro-
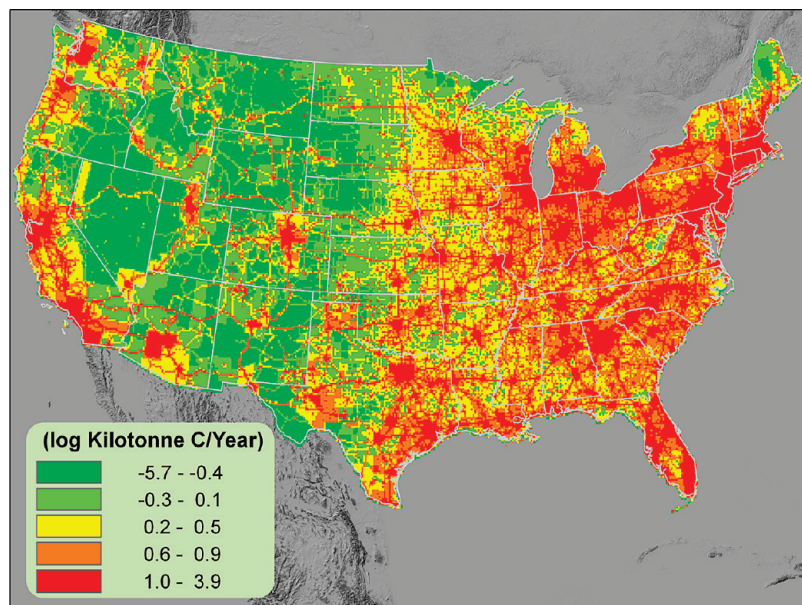
FIGURE 2. Total contiguous U.S. fossil fuel CO_2 emissions for the year 2002 represented on a 10 km × 10 km grid (units: $\log_{10}$ GgC 100 km^{-2} year^{-1}).
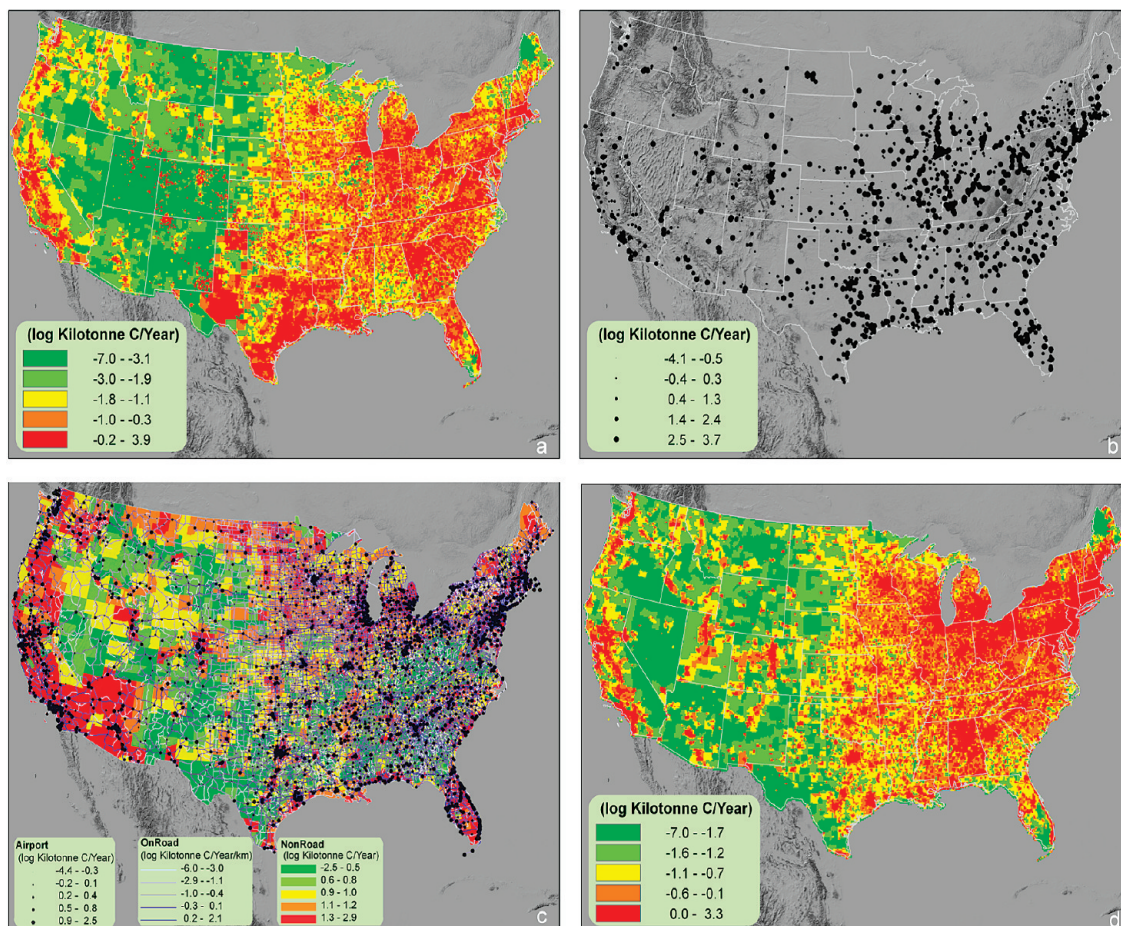

FIGURE 3. Contiguous U.S. fossil fuel CO_2 emissions from the (a) industrial sector ($\log_{10}$ GgC 100 km^{-2} year^{-1}); (b) the electricity production sector ($\log_{10}$ GgC facility^{-1} year^{-1}); (c) the transportation sector ($\log_{10}$ GgC km^{-1} year^{-1} for onroad, $\log_{10}$ GgC facility^{-1} year^{-1} for air transport, $\log_{10}$ GgC 100 km^{-2} year^{-1} for nonroad); (d) the sum of the commercial and residential sectors ($\log_{10}$ GgC 100 km^{-2} year^{-1}).

duced or supplied to the market. At the national/annual scale, very small differences might be expected to occur when comparing the DOE/EIA surveys to the Vulcan estimates. For example, the DOE/EIA does require balancing items in order to balance the supply and consumption of natural gas, and these can be on the order of 0.5 to 1% (*20*). Furthermore, supply and ultimate combustion of fuel may be different due to changes in stockpiling from one year to the next and assumptions about noncombustion oxidation of fossil fuels. This is particularly relevant for liquid petroleum fuel.
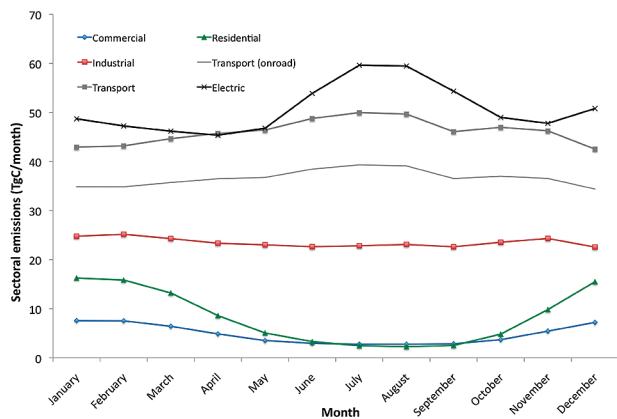
FIGURE 4. 2002 monthly total fossil fuel CO_2 emissions by major economic sector (TgC/month).

FIGURE 5. Annual mean, atmospheric CO_2 concentration difference (Vulcan minus Brenkert) at 850 mb in units of ppmv as generated by the difference between simulated Vulcan fossil fuel CO_2 emissions and simulated Brenkert emissions.

TABLE 1. 2002 U.S. Total Fossil Fuel CO_2 Emissions by Economic Sector As Estimated by the DOE/EIA (9), the EPA (10), and the Vulcan Study[a]

sector	DOE/EIA	Vulcan	% diff	EPA	% diff
commercial	62.4	65.4	4.8		
industrial	289.1	285.5	−1.2		
residential	100.2	100.6	0.4		
mobile	515.1	512.8	−0.5	516.3	−0.7
onroad		399.7		379.1	5.4
nonroad		49.8		72.6	−31.4
aircraft		63.9		64.6	−1.1
electric	616.3	616.9	0.1		
total	1583.1	1581.1	−0.1		

[a] The percent difference between the DOE/EIA estimate and the Vulcan estimate is included as is the difference between the EPA mobile sector estimate and the Vulcan estimate. Units: TgC year^{-1}.

The largest discrepancy is in the transportation sector, where Vulcan emissions for the nonroad sector are 31.4% lower than the EPA estimate. However, because the transportation sector is dominated by the onroad component, the overall match is within 1%. The nonroad discrepancy is due to the lack of railroad and commercial marine vessels in the Vulcan inventory. This data was not present in the NMIM NCD. This missing component will be included in future versions.

Comparison to Previous Gridded Inventory. A critical goal of the Vulcan research is to contribute to improved quantification of the North American carbon budget, particularly for scales equivalent to, or finer than, the U.S. county level. Because of the accuracy and integrating ability of atmospheric measurements, the total carbon budget is often explored though a combination of surface flux estimation and atmospheric measurements. Hence, we compare the atmospheric concentration of CO_2 resulting from the Vulcan inventory to the same derived from the previous inventory used in carbon cycle research. It is worth noting that the use of atmospheric measurements to evaluate emission inventories remains an imperfect tool due to the fact that the atmospheric approach must rely on simulation of atmospheric transport, an effort that remains a substantial research item.

The high resolution fossil fuel emissions data product, generated by A. Brenkert (http://cdiac.ornl.gov/epubs/ndp/ndp058a/ndp058a.html), widely used in carbon cycle studies, was a 1° × 1° emissions product based on 1995 national-level fossil fuel consumption combined with population density (7, 8). The Brenkert inventory was subsampled to 0.1° × 0.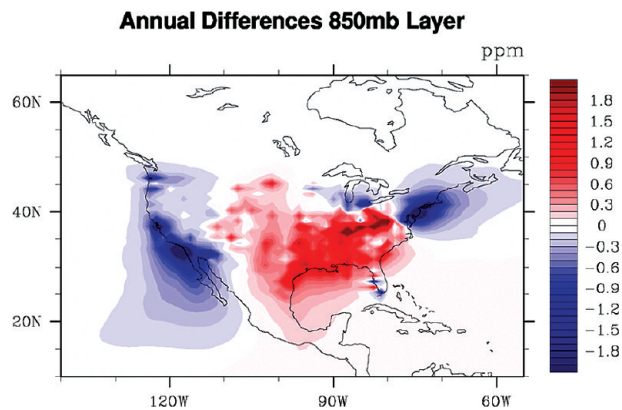1° and scaled to match the national total of the Vulcan inventory. A difference map was created by subtracting a 0.1° × 0.1° Vulcan flux field from the scaled Brenkert inventory [see Supporting Information, SI Text 2 and SI Figure 4]. The Vulcan inventory estimates higher emissions in the Southeast and along portions of the Texas Gulf coast and less emissions in the Northeast and upper Midwest. Some of this may be due to population migration from 1995 to 2002, but it is more likely due to the decoupling of emissions and population. For example, the gas and oil production industry is a significant proportion of the Gulf coast emissions and are often not coincident with large population centers. This is most obvious for electricity production and some industrial sectors which are often not collocated with high population centers.

Calculated covariance between the Vulcan sectors and the population-based Brenkert inventory show the Vulcan electricity production sector having the smallest spatial covariance (0.072) while the Vulcan commercial and residential sectors the highest (0.65 and 0.35, respectively), consistent with expectations that large point sources are increasingly sited away from population centers while commercial and residential emitters are typically collocated with population centers.

Figure 5 shows the difference in annual mean CO_2 concentration at 850 mbar resulting from the Vulcan versus Brenkert inventories via forward integration of the Parameterized Chemical Transport Model (PCTM) (21). Concentration differences as large as 2 ppmv occur associated primarily with the fact that a significant amount of the emissions in the Vulcan inventory are not associated with centers of high population density, the assumption underlying the 1° × 1° inventory. Because both inventories in this simulation experiment have the same U.S. total, the differences in Figure 5 reflect differences in spatial allocation of the U.S. total. By way of context, model simulated interhemispheric differences arising from fossil fuel fluxes are roughly 3 to 4 ppmv at the surface (22). Furthermore, a well-mixed emission of 1 PgC year^{-1} increases the atmospheric CO_2 concentration by ~0.4 ppmv at steady state. Hence, the concentration differences noted in Figure 5 portend significant implications for carbon cycle research.

Caveats. The Vulcan inventory has key caveats associated with the current data product release:

• The data sources for the emissions estimates were, with minor exceptions, taken "at face value" with no further QA/QC beyond what the providing agencies perform.

• In terms of introduced uncertainty, assignment of non-CO_2 pollutant emission factors and estimates of vehicle miles traveled contain the greatest amount of uncertainty and

unaccounted-for variability. Though formal uncertainty has not been presented here, extensive uncertainty and sensitivity analysis is underway and will accompany future releases of the Vulcan data product. Preliminary sensitivity tests have concluded that two sigma errors could be as high as 16% at the state level and 50% at the county level.

• The downscaling of the nonpoint sources relies on the use of building square footage. Other predictive factors not considered in the subcounty downscaling would be variables such as building age, occupancy, and nonspace heating share of fuel use.

• The submonthly temporal downscaling of the mobile emissions remains limited in terms of capturing true spatial variability.

• The 48 fuels tracked in the Vulcan inventory are represented by U.S.-average heat contents, carbon contents, and carbon fraction oxidized. This overlooks potential spatial and temporal variation in these parameters.

Implications. The Vulcan inventory has wide-ranging implications for carbon cycle and climate change research and utility for national legislation on climate change and energy policy. As the largest net land−atmosphere flux, fossil fuel CO_2 plays a key role in atmospheric CO_2 inversions, widely used to identify and understand the net terrestrial flux. Space/time biases in the fossil fuel flux are directly aliased into the net terrestrial flux in these studies (*4*). As inversion studies move to smaller space/time scales, the fossil fuel CO_2 flux must be accurately quantified at smaller space/time scales. This is particularly true as remotely sensed CO_2 observations become available from the GOSAT mission.

High resolution, process-driven inventories also make possible the incorporation of much finer, process-driven emissions into integrated assessment studies which currently utilize spatially coarse emission projections driven from indirect measures such as population and economic growth.

Finally, high resolution inventories can provide a carbon trading system with a single, science-driven inventory platform. This could widen the scale and sectoral detail for carbon trading. It could also offer a method by which progress in emissions mitigation can be tracked and performance confirmed. This last point is crucial in that currently there is no systematic way to evaluate the progress of mitigation measures, an essential component of assessing and judging success as the U.S. constructs legislative measures on climate change mitigation.

Future research direction includes quantifying emissions from Canada and Mexico in order to complete the entire North American inventory. Work has also begun on comparison to recently completed state/month level inventories built from fuel sales and supply (*13, 9*). The 10 km × 10 km Vulcan inventory for each of the sectoral contributions has recently been placed within the Google Earth Platform, and further work is planned to provide more of the detail within the complete Vulcan data product such as a "native" resolution product (geocoded points, roads, etc) and combustion category and fuel types.

Finally, research has begun on building a fossil fuel CO_2 inventory at the scale of individual buildings in near real-time through a combination of in situ measurements, remote sensing, and energy systems modeling. This project, called "Hestia", is currently utilizing the city of Indianapolis as a first pilot location and is complemented by airborne CO_2 flux measurement campaigns from low-flying light aircraft (*23*).

Supporting Information Available

The supporting information contains a more detailed elaboration of the datasets, emissions calculations and downscaling methods employed in this study. This material is available free of charge via the Internet at http://pubs.acs.org.

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