# Improving the temporal and spatial distribution of CO<sub>2</sub> emissions from global fossil fuel emission data sets

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[1] Through an analysis of multiple global fossil fuel CO<sub>2</sub> emission data sets, Vulcan emission data for the United States, Canada's National Inventory Report, and NO<sub>2</sub> variability based on satellite observations, we derive scale factors that can be applied to global emission data sets to represent weekly and diurnal CO<sub>2</sub> emission variability. This is important for inverse modeling and data assimilation of CO<sub>2</sub>, which use in situ or satellite measurements subject to variability on these time scales. Model simulations applying the weekly and diurnal scaling show that, although the impacts are minor far away from sources, surface atmospheric  $CO_2$  is perturbed by up to 1.5-8 ppm and column-averaged  $CO_2$  is perturbed by 0.1–0.5 ppm over some major cities, suggesting the magnitude of model biases for urban areas when these modes of temporal variability are not represented. In addition, we also derive scale factors to account for the large per capita differences in  $CO_2$  emissions between Canadian provinces that arise from differences in per capita energy use and the proportion of energy generated by methods that do not emit  $CO_2$ , which are not accounted for in population-based global emission data sets. The resulting products of these analyses are global  $0.25^{\circ} \times 0.25^{\circ}$  gridded scale factor maps that can be applied to global fossil fuel CO<sub>2</sub> emission data sets to represent weekly and diurnal variability and  $1^{\circ} \times 1^{\circ}$  scale factor maps to redistribute spatially emissions from two common global data sets to account for differences in per capita emissions within Canada.

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# 1. Introduction

[2] There is increasing interest in the estimation of  $CO_2$  sources and sinks by inverse modeling or data assimilation techniques; however, current approaches typically do not optimize estimates of  $CO_2$  emissions from fossil fuel combustion [*Gurney et al.*, 2002, 2004; *Baker et al.*, 2006; *Peters et al.*, 2005, 2007; *Nassar et al.*, 2011; *Takagi* 

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*et al.*, 2011]. Instead, fossil fuel emissions of  $CO_2$  are usually treated as a known quantity in the inversion (i.e., with zero uncertainty), so that  $CO_2$  fluxes from the terrestrial biosphere and oceans, which are considered more uncertain, can be optimized using measurements of atmospheric  $CO_2$ . As a result of this approach, errors in the fossil fuel emissions are hidden and instead cause systematic errors in the biospheric and oceanic  $CO_2$  flux estimates. As more advanced data assimilation systems are developed with the aim of producing  $CO_2$  flux estimates with lower (and better characterized) uncertainties to improve our understanding of carbon cycle science or to inform policy discussions, parallel efforts are required to improve all components of the assimilation system.

[3] Global fossil fuel CO<sub>2</sub> emission data sets are typically developed by spatially distributing emissions from national CO<sub>2</sub> emission totals reported under the United Nations Framework Convention on Climate Change (UNFCCC) or other global compilations [e.g., *Andres et al.*, 2012]. The magnitude of the uncertainty in the national totals is thought to be less than ~8% for the developed countries (Annex B countries under the Kyoto Protocol to the UNFCCC), which have strict emissions accounting systems [*Andres et al.*, 1996]. Uncertainties are greater for the developing countries (non-Annex B countries) [*Guan et al.*, 2012], which

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continue to make an increasing contribution to global CO<sub>2</sub> emissions, now exceeding the emissions from developed countries [Peters et al., 2011]. Errors related to the spatial and temporal distribution in emission data sets are more difficult to quantify but can be proportionally much larger than the national totals. Although these emission data sets typically include interannual variability, which has a major impact on the estimation of annual CO<sub>2</sub> fluxes, the implementation of intra-annual temporal variability is still novel. The gridded  $1^{\circ} \times 1^{\circ}$  emission data set from the Carbon Dioxide Information and Analysis Center (CDIAC) [Andres et al., 2011], which distributes national emissions according to population density, includes global seasonal variability using monthly emission totals for many countries. The Open-Source Data Inventory of Anthropogenic CO<sub>2</sub> Emission (ODIAC) distributes national emissions based on satellite observations of nighttime lights and information on 17,688 power plants [Oda and Maksyutov, 2011] providing emissions at a selection of spatial resolutions  $(1^{\circ} \times 1^{\circ}, 0.05^{\circ} \times 0.05^{\circ}, \text{ or } 0.01^{\circ} \times 0.01^{\circ})$ . A new version of ODIAC (v3.0) includes a seasonal cycle using the CDIAC approach [Maksyutov et al., 2012]. The Emission Database for Global Atmospheric Research (EDGAR; http://edgar.jrc.ec.europa.eu/index.php) version 4.2 distributes national emission totals using a geographic database with data such as the locations of energy and manufacturing facilities, road networks, shipping routes, human and animal population density, and agricultural land use, resulting in  $0.1^{\circ} \times 0.1^{\circ}$  annual emission maps, but EDGAR v4.2 has no intra-annual variability. On the other hand, the Vulcan fossil fuel emission data product [Gurney et al., 2009], derived from a complex mix of data from power plant CO emissions, transportation statistics, and a variety of socioeconomic indicators, gives hourly CO<sub>2</sub> emissions at high spatial resolution  $(10 \times 10 \text{ km}^2 \text{ and } 0.1^\circ \times 0.1^\circ)$ but is limited to the United States for the year 2002. Another regional high-spatial- and high-temporal-resolution data set also exists but includes only U.S. power plants [Pétron et al., 2008].

[4] The absence of temporal variability on the scale of days or weeks in CDIAC and ODIAC, or any intra-annual variability in standard EDGAR emission data, means that nearly all models using them assume constant emissions for those time scales, even though energy use is known to exhibit weekly and diurnal cycles. A set of weekly and diurnal temporal curves was provided as an EDGAR v3.2 provisional data product determined using emissions patterns from Western Europe, as an exploratory method of addressing this problem (http://themasites.pbl.nl/en/ themasites/edgar/documentation/content/Temporal-variation. html). Although the associated disclaimer warns of high uncertainty in applying the temporal curves to other parts of the world, they have been applied to the global  $1^{\circ} \times 1^{\circ}$ EDGAR v3.2 emission data set with few modifications as part of the CO<sub>2</sub> Release and Oxygen Uptake from Fossil Fuel Emission Estimate (COFFEE) [Steinbach et al., 2011]. The Institute of Energy Economics and the Rational Use of Energy (IER) at the University of Stuttgart is also actively working in the area of short-time-scale variability in CO<sub>2</sub> emissions from fossil fuels for Europe [Pregger et al., 2007; see also IER Emission Data, 2008 (http://carboeurope.ier. uni-stuttgart.de/)].

[5] The importance of seasonal variability in fossil fuel emissions to CO<sub>2</sub> inverse modeling has been demonstrated in previous work [Gurney et al., 2005; Erickson et al., 2008; Peylin et al., 2011]. The importance of weekly or diurnal variability in CO<sub>2</sub> inverse modeling has received less attention, but, in a multimodel experiment using IER hourly emission data and EDGAR FT2000 data with hourly variability, Peylin et al. [2011] explore the impact of intra-annual variability on CO<sub>2</sub> simulations over Europe. Although Peylin et al. [2011] determined that the impact of the intra-annual variability in emissions is less than the impact of using different transport fields, it was still found to be an important factor, and the authors referred to an urgent need for more work on improving the spatial and temporal distributions of fossil fuel emission data. One important reason for including weekly or diurnal variability is the fact that the measurements used in inverse modeling studies are subject to these temporal scales. CO2 flask samples are commonly collected only once or twice per week and correspond to a specific time of the day (usually afternoon) [Conway et al., 1994]. The use of continuous in situ CO<sub>2</sub> measurements (which span the diurnal and weekly cycles) is also becoming more common [Worthy et al., 2003]. With the arrival of novel satellite observations of CO<sub>2</sub> from missions such as the Greenhouse Gases Observing Satellite (GOSAT) [Yokota et al., 2009; Yoshida et al., 2011] and the upcoming Orbiting Carbon Observatory 2 (OCO-2) mission [Crisp et al., 2004, 2012], new, advanced data assimilation systems are being developed to take advantage of these data. As additional CO<sub>2</sub> satellites enter into operation and the data assimilation systems mature further, they may eventually be used for international treaty monitoring and verification purposes [Pacala et al., 2010]. Polar-orbiting nadir-viewing satellites (such as GOSAT and OCO-2) usually have sun-synchronous orbits, which have a fixed equator-crossing time and thus provide observations at a set local time of the day. Satellite observations of NO<sub>2</sub> from such orbits have already demonstrated clear variability in anthropogenic emissions over the course of a week or the diurnal cycle [Beirle et al., 2003; Boersma et al., 2009]. Because CO<sub>2</sub> has a longer atmospheric lifetime than NO<sub>2</sub>, short-time scale variability will have less of an impact on CO<sub>2</sub> because enhancements relative to background levels are smaller, but accounting for this variability in advanced CO<sub>2</sub> data assimilation systems (aimed at making more accurate estimates) could reduce errors in the optimization of CO<sub>2</sub> fluxes from other sources and sinks.

[6] In this work, we have developed a set of seven scale factor maps for each day of the week (weekly) and 24 scale factor maps for each hour of the day (diurnal) at  $0.25^{\circ}$  0.25° resolution globally. We refer to this new data product as Temporal Improvements for Modeling Emissions by Scaling (TIMES). The TIMES weekly- and diurnal-scale factor maps can be applied to global CO<sub>2</sub> emission data sets lacking these modes of temporal variability, such as CDIAC, ODIAC, EDGAR, and others, to reduce errors in 3D CO<sub>2</sub> modeling or data assimilation. This study was motivated by the need to provide accurate fossil fuel emissions for an advanced global CO<sub>2</sub> data assimilation system now under development at Environment Canada, which will use both in situ and satellite observations of CO<sub>2</sub>, will apply data assimilation methods from numerical weather prediction, and



**Figure 1.** Dimensionless hourly scale factors for the five sectors with diurnal variability (RES, residential; IND, industrial; UTL, electric utilities; COM, commercial; MOB, on-road mobile) in the Vulcan emission data product, along with the weighted total (TOT; which also includes the remaining emissions) for the contiguous United States in 2002. These were calculated by averaging the temporal profiles from the four time zones of the contiguous United States with the necessary offset. MOB has the largest diurnal amplitude of all sectors, whereas RES and COM are almost indistinguishable, with small diurnal amplitudes.

will attempt to account for all sources of uncertainty. This system is still being developed, so we test the sensitivity of surface atmospheric CO<sub>2</sub> distributions to these temporal-scale factors with the GEOS-Chem model [*Nassar et al.*, 2010]. In the process of developing the temporal-scale factors, limitations in current methods of spatially distributing emissions were also highlighted. One key limitation relates to the wide range of variability in per capita emissions that can exist within a country, which is not accounted for explicitly in population-based inventories; therefore, as an additional data product, a separate set of  $1^{\circ} \times 1^{\circ}$  Canadian scale factor maps was developed that could be applied to the CDIAC and ODIAC emission inventories to improve the spatial distribution of Canadian CO<sub>2</sub> emissions.

# 2. Methods

## 2.1. Diurnal Variability

[7] Our approach to developing the TIMES scale factor maps begins with an analysis of the Vulcan emission data product v2.0, which provides CO<sub>2</sub> emissions for the contiguous United States in 2002 at  $10 \times 10 \text{ km}^2$  and  $0.1^\circ \times 0.1^\circ$  resolution. (The v2.2 update also includes Alaska at  $10 \times 10 \text{ km}^2$ .) Vulcan emissions are divided into eight sectors: residential, commercial, industrial, electricity production, mobile on-road, mobile nonroad, cement manufacture, and aircraft [*Gurney et al.*, 2009]. The residential, commercial, industrial, electricity productors exhibit diurnal variability, whereas the other sectors are temporally constant in Vulcan at the diurnal scale (although they have monthly time structure).

[8] Diurnal variability in anthropogenic  $CO_2$  emissions is related to both geophysical cycles (i.e., heating/cooling needs of a building changing with diurnal cycle in outdoor temperatures) and sociodemographic patterns, such as the time of day when people drive to/from work. Representing the geophysical contribution to the diurnal cycle could perhaps be accomplished best by relating emissions to the local solar time (which varies continuously as a function of longitude): however, representing the sociodemographic aspect of diurnal variability would suggest the use of standard international time zones, which are essentially a stepwise representation of solar time. Therefore, we determined the diurnal cycle for emissions from each Vulcan sector separately for the four time zones in the contiguous United States. Averaging the results from the different time zones and accounting for their hourly offsets (neglecting daylight saving time) gives the United States diurnal profiles for these sectors shown in Figure 1, where a value of 1 represents the daily mean for each sector. The profile for total emissions shown in Figure 1 is dependent on the fraction that each sector contributes to the total (sector weighting).

[9] Based on Vulcan for the United States (at  $0.1^{\circ} \times 0.1^{\circ}$ ) and EDGAR v4.2 for other countries, the proportion of the emissions from the residential, industrial, electricity production, mobile on-road, and "other" sectors was determined for the 20 highest CO<sub>2</sub>-emitting countries in 2008 (Table 1), which account for 80% of global CO<sub>2</sub> emissions from fossil fuel combustion and cement manufacture [Andres et al., 2011]. The diurnal profiles for each sector from Vulcan were each associated with a sector from EDGAR, then weighted according to the EDGAR sector proportions to determine a diurnal profile of total CO2 emissions for the other 19 high-emitting countries. The remaining countries of the world were each associated with a proxy country from the top 20 CO<sub>2</sub>-emitting countries to approximate their diurnal variability. Figure 2 shows these proxy country groups; for example, the diurnal variability of Portugal was based on

**Table 1.** Fraction of Emissions per Source Sector for the 20 Highest-Emitting Countries in 2008 From EDGAR v4.2 and for theContiguous United States in 2002 From Vulcan<sup>a</sup>

Country	Residential	Industrial	Utilities	Mobile	Other	
China	0.067	0.368	0.494	0.046	0.025	
United States	0.109	0.142	0.420	0.278	0.050	
India	0.108	0.238	0.552	0.081	0.022	
Russia	0.116	0.201	0.528	0.116	0.040	
Japan	0.129	0.246	0.426	0.163	0.036	
Germany	0.198	0.155	0.416	0.178	0.053	
Canada	0.158	0.204	0.303	0.241	0.094	
Iran	0.346	0.398	0.197	0.025	0.034	
United Kingdom	0.183	0.144	0.382	0.221	0.071	
South Korea	0.107	0.213	0.493	0.151	0.036	
Mexico	0.075	0.244	0.332	0.320	0.029	
Italy	0.170	0.213	0.309	0.249	0.058	
South Africa	0.077	0.153	0.621	0.130	0.018	
Saudi Arabia	0.013	0.230	0.391	0.300	0.066	
Indonesia	0.071	0.357	0.363	0.158	0.050	
Australia	0.040	0.144	0.608	0.166	0.042	
Brazil	0.093	0.380	0.123	0.356	0.049	
France	0.226	0.223	0.157	0.306	0.088	
Spain	0.097	0.217	0.301	0.299	0.086	
Ukraine	0.136	0.313	0.412	0.100	0.039	
U.SVulcan	0.066	0.188	0.400	0.258	0.088	

<sup>a</sup>The four sectors shown have short-time scale variability in Vulcan and are also present in EDGAR. "Other" is the sum of sectors that did not have short-time scale variability in Vulcan. The "Other" value for Vulcan also includes the commercial sector, which does have short-time scale variability but is not defined in EDGAR.



**Figure 2.** Twenty proxy country groups for weekly and diurnal scale factors, for which each group is associated with one of the 20 highest  $CO_2$ -emitting countries (Table 1). The highest-emitting countries were based on the CDIAC ranking for 2008, but these groupings differ from the 21 proxy country groups used by *Andres et al.* [2011].

that of Spain, New Zealand was based on Australia, and all African nations were based on South Africa. The approach of using of the highest-emitting countries as proxies was also applied in the CDIAC emission product to obtain monthly variability [Andres et al., 2011]. Although our proxy country groups sometimes differ from those of Andres et al. [2011], their work showed that the sensitivity to proxy country selection was low (for the global seasonal cycle), partially because the emissions from nonproxy countries are relatively low. Canadian provincial and territorial energy sector information is readily available in Part 3 of Canada's National Inventory Report [NIR, 2011], so we treated each Canadian province and territory separately, using these factors (Table 2) instead of the EDGAR values for Canada. Canadian provincial differences and the National Inventory Report are discussed in detail in section 2.3.

[10] An open-source time zone shape file was gridded (using Geographic Information Systems [GIS] software) to produce a  $0.25^{\circ} \times 0.25^{\circ}$  global time zone map. The 399 time zone shapes were then grouped into 24 time zones, essentially rounding off time zones with offsets of less than 1 h and aggregating time zone shapes with the same standard time but different daylight saving time rules. A dimensionless scale factor from the diurnal profile of total  $CO_2$  emissions for each country was then gridded to  $0.25^{\circ}$ 0.25° using country and provincial masks at this resolution to give global scale factors maps for each hour of the day, with appropriate offsets for time zones. Figure 3 shows global diurnal scale factor maps for 00, 06, 12, and 18 Coordinated Universal Time (UTC). These dimensionless factors can be applied to scale emissions from an emission data set without variability in total emissions over a 24 h period. The factors have six or more significant digits to ensure conservation of mass for total emissions after scaling, not to imply that true values are known to this precision.

[11] The sensitivity of surface atmospheric  $CO_2$  to the diurnal scale factors was determined using the GEOS-Chem model's CO<sub>2</sub> simulation mode [*Nassar et al.*, 2010]. Version 9.01.01 of the model was run using GEOS-5 meteorology for 2009 at  $2^{\circ} \times 2.5^{\circ}$  horizontal resolution with 47 vertical hybrid sigma-pressure levels from the surface to 0.01 hPa. The  $1^{\circ} \times 1^{\circ}$  CDIAC fossil fuel and cement CO<sub>2</sub> emission data set for 2007 was used [*Andres et al.*, 2011]. Sensitivity was determined by taking the difference between a simulation that had scale factors applied and a simulation without them (with identical initial conditions and fluxes), yielding CO<sub>2</sub> perturbations ( $\Delta$ CO<sub>2</sub>). Maps of  $\Delta$ CO<sub>2</sub> are also shown in Figure 3, illustrating the spatial impacts, but, because some pixels are saturated using the selected color scale, the maximum and minimum values are not evident and instead

**Table 2.** Fraction of Emissions by Sector for Canadian Provincesand Territories in 2009 From Canada's National Inventory Report,Part 3 [2011]<sup>a</sup>

Province	Residential <sup>b</sup>	Industrial	Utilities	Mobile	Other <sup>c</sup>
British Columbia (BC)	0.140	0.193	0.024	0.290	0.353
Alberta (AB)	0.077	0.237	0.261	0.113	0.313
Saskatchewan (SK)	0.081	0.122	0.327	0.152	0.318
Manitoba (MB)	0.213	0.176	0.014	0.438	0.159
Ontario (ON)	0.217	0.224	0.108	0.312	0.139
Quebec (QC)	0.169	0.215	0.008	0.418	0.190
New Brunswick (NB)	0.071	0.094	0.411	0.215	0.210
Nova Scotia (NS)	0.133	0.038	0.500	0.179	0.150
Prince Edward Island (PE)	0.337	0.055	0.000	0.466	0.142
Newfoundland and	0.095	0.108	0.104	0.245	0.449
Labrador (NL)					
Territories (Ter)	0.157	0.189	0.020	0.198	0.436
Canada	0.136	0.202	0.179	0.235	0.248

<sup>a</sup>Three significant digits are shown for simplicity, but more were used in the actual calculation.

<sup>b</sup>Residential here is actually the sum of residential and commercial.

<sup>cu</sup>Other" from the NIR accounts for a larger contribution than in EDGAR because of differences in the definitions of the sectors, related primarily to electric utilities.





**Figure 3.** (Left) Global  $0.25^{\circ} \times 0.25^{\circ}$  dimensionless diurnal scale factor maps for 00, 06, 12, and 18 UTC. The boundaries between colored regions coincide with time zone boundaries, national borders, or Canadian provincial borders. Regions with a high proportion of emissions from on-road transportation have larger diurnal amplitudes. Both the minimum and the maximum can be seen to move westward throughout the course of a day. (Right) The CO<sub>2</sub> perturbation ( $\Delta$ CO<sub>2</sub> determined as scaled – control) in the lowest atmospheric layer that results from applying the diurnal scale factors in a model for a typical December day. Because of the time lag of a few hours for the maximum atmospheric impact of the emissions scaling, the right panels are offset by 6 hours to obtain the best correspondence with the left panels. Impacts are largest over the highest-emitting regions (Eastern United States, Europe, and China), but, because some pixels are saturated (outside the color scale range) to illustrate the spatial effects better, the full magnitude of the impact in those regions cannot be determined accurately from this figure.

are dealt with in the Discussion. Offsetting the sensitivity maps by 6 h in Figure 3 gives a better match between the scaling factors and the  $\Delta CO_2$  perturbation in the atmosphere, which emphasizes that there is a time lag of a few hours between the most intense scaling and the maximum atmospheric perturbation, but the exact time lag will vary based on local transport factors.

#### 2.2. Weekly Variability

[12] The same sectors with diurnal variability in Vulcan also have weekly variability. For these sectors, emissions on Saturday and Sunday are generally lower than on weekdays, with the greatest differences seen for on-road mobile followed by industrial emissions. Dimensionless factors were therefore determined for each sector on Saturday, Sunday, and weekdays, in which 1 corresponds to the weekly mean. These individual sector factors were used to calculate a sector-weighted total factor yielding 0.913268 for Sunday, 0.962927 for Saturday, and 1.024760 for weekdays, as shown in Table 3. A quick comparison with the diurnal scale factors from Figure 1, indicates that the scale factors for the weekly cycle are closer to 1 than the diurnal cycle.

[13] The reason for a weekly cycle in  $CO_2$  emissions is entirely sociodemographic rather than geophysical and relates

	Residential	Commercial	Industrial	Utilities	Mobile	Total
Weekday	1.004859	1.003891	1.051292	1.026384	1.022697	1.024760
Saturday	0.984441	0.989275	0.873448	0.942249	1.023480	0.962927
Sunday	0.991269	0.991267	0.870090	0.925828	0.882269	0.913268

**Table 3.** Weekday and Weekend Scale Factors for Five Sectors and the Weighted Total From Vulcan for the Contiguous United States in 2002<sup>a</sup>

<sup>a</sup>The weekly mean has a value of 1.

to the customs and commercial schedules of the local population and thus will differ for different parts of the world. Different weekend definitions were not accounted for in the COFFEE implementation of weekly cycles [Steinbach et al., 2011], but satellite observations of the vertical column density of NO<sub>2</sub> (a short-lived tracer closely linked to anthropogenic emissions), averaged over the period 1996-2001 show different weekly cycles over many major cities of the world [Beirle et al., 2003]. U.S. and European cities, Mexico City, and others exhibited their primary minimum in NO<sub>2</sub> on Sunday, with slightly lower values on Saturday relative to the rest of the week, consistent with the traditional nonworking days in the West (related to Christian customs), as was seen in the Vulcan CO2 emissions. For regions with other dominant religions, the weekly cycle was not as clear. NO<sub>2</sub> variability over Japanese and South Korean cities resembled that in the West, but many Middle Eastern cities (Cairo, Riyadh, Mecca, Abu Dhabi) had minimum NO<sub>2</sub> on Friday, consistent with their main day of rest, although Jerusalem had a minimum on Saturday. Over the past decade, numerous Middle Eastern countries have redefined their weekend from Thursday-Friday to Friday-Saturday, in order to balance the desire to synchronize their economies with their neighbors and the norms of international trade and yet respect Islamic religious traditions. Gavison and Perez [2007] provide a discussion on days of rest in various areas of the world. An updated online list of days of rest for various countries (http://en.wikipedia.org/wiki/Workweek and weekend) was used in this work.

[14] In the work of Beirle et al. [2003], Chinese cities did not show any weekly NO2 cycle discernible from the noise, which was attributed to the fact that Chinese NO<sub>x</sub> emissions were dominated by power plants and heavy industry operating throughout the entire week, whereas in many other parts of the world transportation emissions play a larger role. Although CDIAC (2011 update) indicates that China's CO<sub>2</sub> emissions doubled from 0.95 PgC in 2001 to 1.92 PgC in 2008, the proportion of emissions from road transportation has held roughly constant (~4%) over this period according to EDGAR v4.2 (road transportation emissions contribute  $\sim 26\%$  in the United States according to Vulcan). However, Table 3 shows that industrial emissions make the largest contribution to the U.S. weekly cycle, so perhaps the lack of a weekly cycle in China's NO2 relates to different industrial patterns during the time frame of that study.

[15] A weekly cycle in total CO<sub>2</sub> emissions for the 20 highest-emitting countries was determined by a weighted application of the U.S. scale factors for the four Vulcan sectors with weekly variability (and a constant factor of 1 for other emissions) to the proportion of the sector contribution to total emissions in EDGAR v4.2 (although the original Vulcan sector weighting is used for the United States). Countries not on the top 20 list were associated with a proxy

country as in the previous section. In some cases, this meant that the weekly scaling was shifted by 1-2 days from the proxy country, as in the case of Syria with a Saturday–Sunday weekend and its proxy Saudi Arabia with a Thursday–Friday weekend. Once again, Canadian provinces were each treated individually using the sector information from the *NIR* [2011] rather than the pan-Canadian value from EDGAR.

[16] All scale factors from this method were then gridded to  $0.25^{\circ} \times 0.25^{\circ}$  using country and provincial masks at this resolution to give global scale factor maps for each day of the week (although Monday-Wednesday are identical), as shown in Figure 4. The sensitivity of surface atmospheric  $CO_2$  to the weekly scale factors was determined using GEOS-Chem by an approach similar to that used in the previous section; however, here the sensitivity is determined using daily average CO<sub>2</sub> instead of an instantaneous value. This avoided selecting a single universal time for each map, since a single time obscures the spatial impacts of the weekly scaling by the diurnal cycle in meteorology, which systematically differs with longitude at a given time. Maps of the  $\Delta CO_2$  from the daily averages are shown in Figure 4, with some pixels saturated using the selected color scale range. These maps show more widespread, but less intense, perturbations than the diurnal impact maps. The maximum intensity of the perturbations is reviewed in the Discussion.

## 2.3. Variability in Per Capita Distributions for Canada

[17] The gridded CDIAC emission data set [Andres et al., 2011] uses population for spatially distributing emissions within a country. Although this may be reasonable for many countries, the reliability of this approach decreases for larger countries (and finer spatial scales), because regions could differ widely in terms of their energy-intensive industries, electricity generation methods, transportation patterns, heating/cooling requirements, or even overall standard of living. Per capita differences in the United States were identified by Pétron et al. [2008]. Zhou and Gurney [2011] demonstrate deviations from the national mean per capita CO<sub>2</sub> emissions for the United States based on Vulcan, and Gurney et al. [2009] show the impact of these differences on an atmospheric CO<sub>2</sub> simulation, indicating annually averaged differences of up to  $\pm 1.8$  ppm at 850 hPa relative to a simulation with a constant per capita emissions. The most densely populated parts of the United States, particularly the West Coast, East Coast northeast from Washington D.C., and the area of Chicago and environs, were associated with lower atmospheric CO<sub>2</sub> in the simulation using Vulcan (indicative of per capita emissions below the U.S. mean), whereas most other parts of the United States had higher CO<sub>2</sub> concentrations. Other analyses comparing modeled atmospheric CO<sub>2</sub> based on Vulcan emissions relative to a simulation using a uniform per capita emission distribution [Corbin et al., 2010, Schuh et al., 2010] were in general agreement with



**Figure 4.** (Left) Global  $0.25^{\circ} \times 0.25^{\circ}$  dimensionless weekly scale factor maps for Sunday, Monday Wednesday, Thursday, Friday, and Saturday, accounting for the different definitions of the weekend around the world. (Right) The daily-averaged CO<sub>2</sub> perturbation ( $\Delta$ CO<sub>2</sub>) in the lowest atmospheric layer that results from applying the weekly scale factors in a model for a typical December week. As with the diurnal scale factors, the impacts are greatest over the high-emitting regions (where some pixels are saturated), but the impacts of the weekly variability are less intense and more dispersed.

the earlier result but also showed localized differences of as much as  $\pm 5.0$  ppm, most often near urban areas.

[18] Canada is the world's second largest country, with an area of  $9.98 \times 10^6$  km<sup>2</sup> and the seventh highest fossil fuel CO<sub>2</sub>-emitting country (according to CDIAC for 2008). It is responsible for ~2% of global emissions, so misrepresenting the spatial distribution of fossil fuel CO<sub>2</sub> emissions within Canada could result in major spatial biases in global model simulations, which would be less significant for smaller countries. Such CO<sub>2</sub> emission biases could impact inverse modeling estimates of terrestrial biospheric CO<sub>2</sub> fluxes. Canada's *NIR* [2011] provides information on greenhouse gas emissions from each province, each province's population, and the breakdown of contributions from different sectors and subsectors (Part 3, Appendices), but the emission values are not distributed spatially for use in

atmospheric CO<sub>2</sub> modeling. In  $1^{\circ} \times 1^{\circ}$  province area masks, the fraction of emissions from each province in the gridded CDIAC (2011 update) and ODIAC v3.0 data relative to the national total (excluding emissions from international shipping and aviation) was determined and compared with the NIR provincial fractions (excluding emissions from agriculture, waste, or land use and land use change from the calculation). Provincial populations and their contribution to national emissions from the NIR, along with scale factors determined from comparing NIR/CDIAC and NIR/ODIAC, are shown in Table 4. Table 5 shows the annual per capita provincial CO<sub>2</sub> emissions from various sectors and subsectors identified in the *NIR* [2011]. Canada's national per capita CO<sub>2</sub> emissions are 4.39 Mg C person<sup>-1</sup> yr<sup>-1</sup>, with Alberta and Saskatchewan well above the national mean (13.68 and 11.94 Mg C person<sup>-1</sup> yr<sup>-1</sup>, respectively), and

Province	Population (thousands)	Fraction of Canadian population	Fraction of Canadian emissions	NIR/CDIAC	NIR/ODIAC	
British Columbia (BC)	4,460	0.132266	0.094649	0.86865	1.38346	
Alberta (AB)	3,671	0.108867	0.339483	3.77577	1.18719	
Saskatchewan (SK)	1,029	0.030516	0.083026	2.26296	1.16052	
Manitoba (MB)	1,220	0.036180	0.021218	0.52631	0.433737	
Ontario (ON)	13,065	0.387456	0.256458	0.68262	0.916361	
Quebec (QC)	7,828	0.232147	0.116052	0.45671	0.685165	
New Brunswick (NB)	749	0.022212	0.030996	0.94641	0.367976	
Nova Scotia (NS)	939	0.027847	0.057934	1.28426	1.70118	
Prince Edward Island (PE)	141	0.004181	0.002362	0.30150	1.070118	
Newfoundland and Labrador (NL)	508	0.015065	0.015240	0.66289	1.29663	
Territories (Ter)	110	0.003262	0.003229	1.12138	0.519725	
Canada	33,720	1	1	0.93040 <sup>b</sup>	0.90035 <sup>b</sup>	

**Table 4.** Population for Canadian Provinces and Territories Along With the Fraction of National Emissions That the Province Contributed in 2009, Determined From Part 3 of Canada's *National Inventory Report* [*NIR*, 2011]<sup>a</sup>

<sup>a</sup>Also shown are scale factors that could be applied to CDIAC and ODIAC to match these values, determined as the NIR fraction divided by the 2008 CDIAC or ODIAC fraction.

<sup>b</sup>Canada ratios differ from 1 primarily because 2009 NIR values were used with 2008 CDIAC and ODIAC values, and Canada's CO<sub>2</sub> emissions declined in 2009. Using 2008 NIR values gives NIR/CDIAC = 0.99065 and NIR/ODIAC = 0.95866, where the remaining difference from 1 likely is due to defining the Canada – United Stattes border with a mask of finite horizontal resolution  $(1^{\circ} \times 1^{\circ})$ .

Quebec, Prince Edward Island, Manitoba, Ontario, and British Columbia below ( $2.19-3.14 \text{ Mg C person}^{-1} \text{ yr}^{-1}$ ), whereas Newfoundland, Nova Scotia, New Brunswick, and the combined territories are near the mean ( $4.34-6.12 \text{ Mg C person}^{-1} \text{ yr}^{-1}$ ).

[19] The high per capita emissions for Alberta and Saskatchewan are the result mainly of the predominant use of coal for the electricity supply (74% and 63%, respectively). However, emissions from fossil fuel production and refining (18.6%) and from oil and gas extraction (12.6%) and fugitive emissions from oil and natural gas (6.2%) in Alberta also play a major role. This is also true for Saskatchewan to a slightly lesser extent (fossil fuel production and refining contributing 13.5%, oil and gas extraction 7.8%), although nonroad mobile emissions (8.7%), presumably related to farming, also contribute at a proportion well above other provinces. Saskatchewan (followed by Alberta) also has the highest per capita emissions from on-road transport according to Table 5. The lower per capita emissions of Quebec and Manitoba arise primarily because 96% and 98% respectively of their electricity was generated using hydroelectricity, which does not directly emit CO<sub>2</sub>. Per capita CO<sub>2</sub> emissions in Ontario were almost as low as those in Quebec and Manitoba, related to the fact that 56% of the Ontario electricity, and 1.5% from nuclear energy, 27% from hydroelectricity, and 1.5% from other renewable energy (wind, solar, geothermal) sources in 2009 [*NIR*, 2011]. The high use of coal and major industrial and

**Table 5.** Per Capita Emissions by Sector (MgC/yr)<sup>a</sup> for Canadian Provinces, the Territories, and the National Mean in 2009, Determined From Part 3 of Canada's National Inventory Report [2011]

	BC	AB	SK	MB	ON	QC	NB	NS	PE	NL	Ter	Can
Energy	3.01	12.94	11.59	2.48	2.61	1.98	6.01	5.58	2.46	4.40	4.34	4.10
a. Stationary Combustion Sources												
Electricity and Heat	0.07	3.57	3.90	0.04	0.32	0.02	2.51	2.82	0.00	0.46	0.09	0.79
Fossil Fuel Production and Refining	0.38	2.55	1.61	0.00	0.12	0.12	0.92	0.37	0.00	0.94	0.06	0.50
Mining & Oil and Gas Extraction	0.11	1.73	0.94	0.03	0.01	0.02	0.02	0.01	0.00	0.42	0.81	0.25
Manufacturing	0.37	0.76	0.14	0.31	0.32	0.23	0.45	0.15	0.12	0.02	0.00	0.34
Construction	0.00	0.01	0.01	0.02	0.01	0.01	0.00	0.01	0.01	0.00	0.00	0.01
Commercial & Institutional	0.18	0.41	0.51	0.32	0.27	0.27	0.26	0.48	0.37	0.28	0.46	0.29
Residential	0.25	0.63	0.46	0.23	0.37	0.10	0.17	0.27	0.47	0.14	0.22	0.31
Agriculture & Forestry	0.00	0.03	0.06	0.01	0.02	0.01	0.01	0.02	0.03	0.00	0.00	0.02
b. Transport												
Domestic Aviation	0.08	0.10	0.05	0.11	0.04	0.03	0.04	0.07	0.03	0.11	0.76	0.06
Road Transport	0.91	1.54	1.81	1.13	0.91	0.92	1.32	1.01	1.16	1.09	0.86	1.04
Rail	0.03	0.21	0.11	0.05	0.02	0.02	0.08	0.04	0.01	0.02	0.00	0.05
Domestic Marine	0.12	0.00	0.00	0.00	0.01	0.04	0.10	0.12	0.14	0.24	0.00	0.04
Off-Road	0.24	0.52	1.03	0.20	0.16	0.17	0.08	0.18	0.14	0.40	1.04	0.25
Pipelines	0.05	0.11	0.59	0.02	0.02	0.01	0.00	0.02	0.00	0.00	0.01	0.05
c. Fugitive Sources												
Oil and Natural Gas	0.20	0.80	0.37	0.01	0.01	0.01	0.06	0.02	0.00	0.28	0.01	0.14
Industrial Processes	0.12	0.74	0.37	0.10	0.31	0.21	0.10	0.04	0.00	0.04	0.01	0.28
Waste	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.02	0.00	0.00	0.00
Total <sup>b</sup>	3.14	13.68	11.94	2.57	2.90	2.19	6.12	5.64	2.48	4.44	4.34	4.39

<sup>a</sup>1 Mg = 1 Tonne

<sup>b</sup>Total is based on the total in the NIR, therefore Total  $\approx$  Energy + Industrial Processes + Waste, but can differ slightly due to the number of significant digits quoted for the total, sector or sub-sector.



**Figure 5.** CDIAC and ODIAC CO<sub>2</sub> emissions for Canada at  $1^{\circ} \times 1^{\circ}$  and the emissions scaled using provincial scale factors (Table 4) based on per capita emissions from Canada's *National Inventory Report* [2011]. Changes in ODIAC are less pronounced than CDIAC, suggesting that nightlights (and the point source list) give a better representation than a purely population-based approach.

agricultural contributions in Alberta and Saskatchewan and the use of non-CO<sub>2</sub>-emitting electricity generation in Quebec, Manitoba, and Ontario highlight a limitation of the population-based distribution of emissions at the national level rather than the provincial level. Application of the scale factors in Table 4 to CDIAC and ODIAC is shown in Figure 5. The fact that the ODIAC nightlights- and point-source-based emission data set has scale factors closer to 1 and changes in Figure 5 are less pronounced suggests that this approach is an improvement relative to CDIAC, but it is still subject to limitations in accounting for emissions from the petroleum industry and farm vehicles or diverted emissions from non-CO<sub>2</sub>-emitting electricity generation. Errors with the locations and the magnitude of point source emissions in ODIAC would not necessarily be highlighted in this approach. The approach used here is most effective for evaluating the province-scale values and provides some suggestion of the validity of the spatial distribution within a province, but it does not evaluate the accuracy of ODIAC spatial distributions at high resolution; however, some of the largest Canadian CO2-emitting point sources located in sparsely populated areas (Fort McMurray, Alberta; Nanticoke, Ontario; and Estevan, Saskatchewan) appear reasonably located in the ODIAC distributions, although they are absent in the CDIAC distributions (Figure 5).

## 3. Results and Discussion

[20] The TIMES scale factor maps developed in this work can be used to represent the diurnal and weekly scale variability in anthropogenic  $CO_2$  emissions by applying them to  $CO_2$  emission data from CDIAC, ODIAC, or EDGAR when used in  $CO_2$  transport models. Previous work has estimated that the diurnal variation resulting from Vulcan emissions can have an impact of nearly  $\pm 3.0$  ppm for select locations, relative to constant daily emissions [*Corbin et al.*, 2010], but in those tests the diurnal effects were not completely isolated from other factors. Direct studies of the diurnal cycle of CO<sub>2</sub> emissions for individual cities have also been conducted [*Vogt et al.*, 2005, *Kühlwein et al.*, 2002], but other work has focused on the change in atmospheric CO<sub>2</sub> concentrations from the diurnal cycle in fossil fuel CO<sub>2</sub> emissions on local scales [*Coutts et al.*, 2007; *Vogel et al.*, 2010].

#### 3.1. Impact on Surface Atmospheric CO<sub>2</sub>

[21] In this section, we investigate the impact of the TIMES scaling factors on surface atmospheric CO2 distributions by taking the difference between simulations with the scaling applied and without. Figures 3 and 4 show that the spatial patterns of the impacts of the diurnal and weekly scaling factors are largest over high-emitting regions, including the Eastern United States, Europe, and China, and moderate over some other regions (U.S. West Coast, the Middle East, India, Japan), but negligible over the majority of the globe. Some pixels are saturated with the chosen color scale (to illustrate better the spatial effects), so the absolute magnitude at a given location within the high-emitting areas is not evident. Figure 6 illustrates the impact of diurnal and weekly scaling over a 60-day period from a GEOS-Chem model simulation, based on two  $2^{\circ} \times 2.5^{\circ}$  surface grid boxes from each of the regions with the largest  $\Delta CO_2$  perturbations, corresponding to New York City, Berlin, and Beijing, as well as two other major cities, Los Angeles and Tehran, from regions of moderate perturbations. The left column shows total CO2 with both temporal scale factors applied



Figure 6

and without any temporal scaling. The differences appear minor relative to the background of about 395 ppm CO<sub>2</sub> and the observed variability, which is mainly related to synoptic scale transport; however, the right column shows the residuals or the  $\Delta$ CO<sub>2</sub> perturbations as a function of time for diurnal scaling, weekly scaling, and combined (weekly + diurnal) scaling, illustrating that the impact is significant at select locations. The contribution from the weekly scaling (~0.5–3.0 ppm) is smaller but more long-lived than the diurnal scaling (~1–5 ppm), as evidenced by the sharper and more regular diurnal fluctuations. Interestingly, the weekly perturbations also display a faint diurnal cycle related to the combination of synoptic transport with higher/lower constant daily emissions.

[22] The combined weekly and diurnal perturbations are characterized by a period of 1 week, in which the most prominent features occur because of the coincidence of the weekly minimum (usually Sunday) with the diurnal minimum and can yield perturbations as large as ~8 ppm for New York City, ~5 ppm for Los Angeles, ~4 ppm for Berlin,  $\sim$ 3 ppm for Beijing and just under  $\sim$ 2 ppm for Tehran. It is presumed that the differences between these locations relate to the absolute magnitude of emissions from the city, the amplitude of the diurnal and weekly scale factors, the local and regional meteorological effects, and the size of the area selected (a  $2^{\circ} \times 2.5^{\circ}$  grid box is  $\sim 222 \times 250 \text{ km}^2$  at these latitudes). Although cities are actually smaller than a single grid box, two boxes were used rather than one because some cities straddle the boundary of two boxes, and a uniform approach was desired for all cities. If a model with higher horizontal resolution was used, the city area could be better defined, likely leading to more intense perturbations.

[23] The combined diurnal and weekly scaling impacts in the range of  $\sim 1.5$  to  $\sim 8$  ppm for large cities of the world can be contrasted with the accuracy (~0.2 ppm) and precision (~0.1 ppm) of flask [Conway et al., 1994] and continuous in situ CO<sub>2</sub> [Worthy et al., 2003] measurements. Both measurement types are commonly used in CO<sub>2</sub> inverse modeling and data assimilation systems, in which the fossil fuel CO<sub>2</sub> is often treated with zero error in order to optimize terrestrial biospheric or oceanic CO2 fluxes. Although these inversions use predominantly background observations to minimize such errors (or inflate the observation errors to account imperfectly for uncertainties like this), as anthropogenic emissions increase, as measurement network coverage expands, and as the desire to constrain fluxes at finer spatial and temporal scales grows, the inclusion of nonbackground sites and reduction of biases from fossil fuel emission inventories will be of greater importance.

#### **3.2.** Impact on XCO<sub>2</sub>

[24] Satellite missions or instruments that measure CO<sub>2</sub> using near-infrared (NIR) or shortwave infrared (SWIR) solar reflectance from the Earth's surface commonly provide a

column-averaged  $CO_2$  mixing ratio referred to as  $XCO_2$ . These observations have vertical sensitivity spanning the entire atmospheric column, including the boundary layer. The sensitivity is nearly uniform vertically throughout the troposphere, then decreases above, with the detailed vertical sensitivity described by a column averaging kernel vector  $a_{CO2}$ [Yoshida et al., 2011]. As a result, the behaviour of XCO<sub>2</sub> differs from that of boundary layer CO<sub>2</sub>, but XCO<sub>2</sub> still contains information about CO<sub>2</sub> near the surface for studying sources and sinks [Olsen and Randerson, 2004]. Satellites in sun-synchronous orbits have a fixed local equator-crossing time for the ascending and descending nodes of each orbit, such that the daylight equator-crossing times are 10:00 for SCIAMACHY [Buchwitz et al., 2007], 12:48 for GOSAT [Yoshida et al., 2011], and ~13:30 for OCO-2 [Crisp et al., 2004, 2012] and the proposed CarbonSat [Bovensmann et al., 2010], so all observations are within minutes of this local time.

[25] Olsen and Randerson [2004] compared the impacts of the biospheric diurnal cycle on surface CO2 and XCO2 by using model simulations. Here, we will compare the impacts of our TIMES fossil fuel scaling on XCO<sub>2</sub> and contrast these results with those of surface  $CO_2$  from section 3.1. Figure 7 shows the perturbations due to the diurnal and weekly scaling factors on XCO<sub>2</sub> calculated using an empirical representation of a mean GOSAT column averaging kernel based on the work of Yoshida et al. [2011]. As in Figures 3 and 4 for surface CO<sub>2</sub>, Figure 7 illustrates the spatial distribution with limited information about the exact magnitude of the impact, because some pixels are saturated with the color scale used. Although the color scale range for XCO<sub>2</sub> has been reduced by a factor of 5 relative to surface CO<sub>2</sub>, the spatial patterns of the perturbations have some similarities, although some important differences are evident too. For example, the relative impact over the north Atlantic due to the US East Coast is more evident for XCO<sub>2</sub> from both diurnal and weekly scaling, and, unlike the case for surface CO<sub>2</sub>, the spatial patterns are slightly more complex, with more frequent "dipoles" consisting of negative and positive lobes in a given region. These dipoles likely result from the transport of air with increased or decreased CO<sub>2</sub> above the surface level near an area where the surface impact has the opposite sign. A qualitative comparison seems to indicate that the impacts from the weekly variability are actually more intense than the diurnal variability, in contrast to the overall case for surface CO<sub>2</sub>, for which the reverse was true. Figure 8 shows the impacts on XCO<sub>2</sub> for the same major cities as in Figure 6, but with the range of the y-axis reduced relative to the surface CO<sub>2</sub> figure. This confirms that the perturbations from weekly variability are typically larger than those from the diurnal variability, which is best illustrated for Beijing.

[26] Although the XCO<sub>2</sub> impacts are generally less than for surface CO<sub>2</sub>, they are significant relative to the 0.3% (~1.0 ppm) precision targeted by OCO-2 [*Crisp et al.*,

**Figure 6.** (Left) Surface-level atmospheric CO<sub>2</sub> at selected major cities with both weekly and diurnal scaling applied (red) and the unscaled control simulation (black) over a 60-day period. (Right) The residuals determined as weekly scaling minus control (red), diurnal scaling minus control (green), and combined temporal scaling minus control (black). These residuals give a measure of the temporally dependent impact or  $\Delta CO_2$  perturbation. The combined impact is highest for New York City, often reaching the range of 5–8 ppm.



**Figure 7.** The column-averaged  $CO_2$  (XCO<sub>2</sub>) perturbation ( $\Delta$ XCO<sub>2</sub> determined as scaled – control) from applying (left) the diurnal scale factors in a model for a typical December day and (right) the weekly scale factors for a typical December week.

2004, 2012] and 0.25 ppm precision of satellite validation data from the Total Carbon Column Observing Network (TCCON) [Wunch et al., 2011]. Hence, we recommend that these modes of temporal variability not be neglected in systems designed for assimilating satellite XCO<sub>2</sub> data. The larger contribution from weekly variability for XCO<sub>2</sub>, in contrast to the situation for surface CO<sub>2</sub>, with the diurnal cycle having a larger impact, could perhaps have been foreseen considering that XCO<sub>2</sub> consists of a vertically integrated signal from air that was in contact with the surface over a larger span of time. It is also consistent with the findings of McKain et al. [2012], who, in an assessment of groundbased CO<sub>2</sub> observations for verification of urban greenhouse gas emissions, state that "integrated column measurements of the urban dome of CO<sub>2</sub> from the ground and/or space are less sensitive than surface point measurements to the redistribution of emitted CO<sub>2</sub> by small-scale processes and thus may allow for more precise trend detection of emissions from urban regions."

## 3.3. Limitations, Uncertainties, and Future Directions

[27] The approach of producing temporal (and spatial) scale factor maps, rather than a new set of emission data files (such as the COFFEE approach) was chosen for this work. The primary reason for this rather than a multiyear, hourly data set is that the scale factor approach provides the variability in modular form, because some users might be interested in per capita scaling but not temporal scaling (or vice versa) or even interested in weekly scaling but perhaps not diurnal scaling (or vice versa).

[28] However, a drawback of the modular scale factor approach is that it makes annual assumptions about the contributions from different sectors, which obviously do change with season. Although the first-order seasonal effect is now accounted for in CDIAC or ODIAC totals, the second-order sector proportional change, which affects the scale factors, is not accounted for in our work, effectively ignoring some of the information available from Vulcan. We have also



**Figure 8.** Same as Figure 6, but for  $XCO_2$  and  $\Delta XCO_2$ . The combined impact is highest for New York City, often reaching the range of 0.4–0.5 ppm.



**Figure 9.** Maps of the standard deviation of the dimensionless diurnal (top) and weekly (bottom) scale factors, which can be used as a measure of uncertainty but do not account for all systematic errors.

applied a single mean diurnal cycle (as mentioned earlier) rather than different weekday and weekend diurnal cycles. Real diurnal cycles for emissions on weekdays and weekends will differ. Weekends are expected to have a largely dampened bimodal (morning/afternoon rush hour) peak in the diurnal cycle because of transportation emissions relative to weekdays and should generally exhibit an increase later in the day relative to the nighttime minimum.

[29] Another significant issue is a temporal bias in the weekly cycle that results for time zones that differ from the zone selected for the day-of-week change. In global modeling work, this will typically be UTC. This problem arises because the weekly scaling map changes at midnight UTC (or another chosen time zone) for the whole globe rather than for the local time. This would be negligible for Berlin, resulting in a phase shift of +1 hour, but the New York weekly cycle would have a phase shift of -5 hours and Beijing +8 hours. For regional modeling (or global modeling with a regional focus), a different time zone could be selected for the day change, which would greatly reduce this effect for the desired locality. However, we note that the diurnal cycle has a larger instantaneous impact than the weekly cycle, where the impact from the weekly cycle is realized as persistently lower weekend emissions over a period of 48 hours, so a cycle with a realistic amplitude, but a phase

shift of a 5-8 hours, is expected to be better than the complete absence of weekly variability.

[30] An alternative approach to deal with the second-order effects (interaction among seasonal, weekly, and diurnal cycles) would be to produce a set of  $12 \times 7$  weekly scale factors or  $12 \times 7 \times 24$  diurnal scale factors to account for these differences, which could be explored in the future. Combining the weekly and diurnal cycles into  $7 \times 24$  maps would remove the phase-shift issue mentioned above. It should also be noted that regridding the scale factors to coarser model horizontal resolution (as in our tests) will result in lowering the amplitude of weekly/diurnal cycles next to regions of no scaling such as oceans, although total emissions will still be properly conserved.

[31] The discussion above highlights the fact that derivation of the TIMES and Canadian spatial scale factors includes a number of approximations and simplifications. The application of more detailed bottom-up methods, such as that used in Vulcan, would likely provide better estimates of CO<sub>2</sub> emissions for high spatial and temporal scales, but these are very labor-intensive undertakings, which to date have been performed for very few countries and only in select years (i.e., United States in 2002). Scale factors allow for immediate application to new releases of inventories such as CDIAC or ODIAC, which occur every year based on annual UNFCCC updates or similar data, with a typical time lag of about 2.5 years (i.e., 2009 emissions distributions are provided no earlier than mid-2012). If a new product were to be produced each time (like COFFEE) instead of scale factors, the additional step required would increase the time lag. The time lag for availability of an emission data product is an issue in the potential application of inventories in future operational  $\hat{CO_2}$  data assimilation systems that could run in near-real time. Additionally, if the objective of such systems was to quantify anthropogenic emissions, using the fossil fuel emission data only as an a priori estimate, this would be consistent with practical approximations such as scale factors.

[32] Uncertainties in the temporal distributions of fossil fuel CO<sub>2</sub> emissions for the EDGAR v3.2 provisional data or COFFEE have not been quantified, nor to date have uncertainties in the spatial distributions of emissions in CDIAC, ODIAC or EDGAR (beyond the country scale), although uncertainties in the spatial distributions have been discussed by Andres et al. [2012]. Quantifying uncertainties in these data products or the scale factors developed here is a challenge. As with the scale factors themselves, their uncertainties will be nonuniform across the globe because of different energy generation methods and usage patterns. A possible method to estimate the uncertainty of the scale factors is to assign the uncertainty related to the level of scaling applied, so, for regions where the scaling amplitude is large, there are larger uncertainties, and, for regions where scale factors are closer to unity, the uncertainties are low. Figure 9 shows maps of the standard deviation of the scale factor values over one cycle (0-23 values for diurnal, 1-7 values for weekly). The  $1\sigma$  uncertainties are from 0.07 to 0.30 for diurnal scaling range and from 0.03 to 0.06 (dimensionless scale units) for weekly scaling. It should be noted, however, that these uncertainty estimates are incomplete, insofar as they relate only to the intensity of scaling and do not account for systematic errors in the method or the input data.

[33] Another approach to estimating uncertainties could be developed later through comparison of simulated  $CO_2$  using the scaling with measurements, perhaps using direct fossil fuel CO<sub>2</sub> tracer methods such as <sup>14</sup>CO<sub>2</sub> [*Turnbull et al.*, 2009; *Levin et al.*, 2011; *Miller et al.*, 2012], because these methods are not subject to the uncertainties in CO<sub>2</sub> fluxes from other components (biosphere, oceans) of the earth system, although they still have their own limitations. It is possible that these studies could identify weaknesses in the current approach, for example, the proxy country groupings, which would require an update to the scale factors, but these changes are expected to be a higher-order correction to the imposition of spatial and temporal variability in emissions that was completely absent in the past.

[34] The use of Canada's NIR for adjusting Canadian emissions for per capita differences is expected to be an improvement upon the current CDIAC and ODIAC emission values, because of the large per capita variation across the country. An evaluation of the sensitivity of atmospheric  $CO_2$  to the spatial scaling, or its impact on  $CO_2$  inversions, will be left for future work. Allocation of emissions per capita is a challenging problem and depends heavily on the spatial scales desired. Although our scale factors now change the relative proportion of the emissions from one province to the next, we have not altered the distribution at finer spatial scales. Hoornweg et al. [2011] indicated that per capita urban emissions are lower than from suburban or rural areas in developed countries because of factors such as urban public transit and smaller dwellings, whereas in developing countries per capita emissions are typically higher in urban areas compared with rural areas and correlate with the general standard of living. That work also showed that, even within a single metropolitan area (or a single neighbourhood), there is a wide range of variability in per capita emissions, but properly accounting for these differences is beyond the scope of this work and is probably better accomplished using bottom-up methods, at least for the time being.

[35] Another promising approach to developing fossil fuel emission inventories with improved spatial and temporal information is the development of a Fossil Fuel Data Assimilation System (FFDAS) [Rayner et al., 2010]. With enough sophistication in such a system, it is theoretically possible to input information to address all of these issues and also estimate uncertainties in emissions, but the current FFDAS is still in its infancy. A modest redistribution of emissions based on per capita differences between provinces, as carried out here, will potentially make a significant difference in continental-scale CO<sub>2</sub> distributions over short time scales, whereas further improvements to represent subprovincial scales better are more challenging, but are expected to have less impact anyway, because of the tendency for sharp pollution gradients to diffuse rapidly, while the transport of larger pollution-enhanced air masses between continents is common.

## 4. Summary and Conclusions

[36]  $CO_2$  emissions from fossil fuel combustion in CDIAC, ODIAC, and EDGAR currently do not include variability in emissions over the course of a week or throughout the day, but inverse modeling and data assimilation systems that use these inventories often deal with measurements

subject to variability on these time scales, such as weekly flask  $CO_2$  measurements or satellite observations of  $CO_2$  from a sun-synchronous orbit corresponding to a single point in the diurnal cycle. Measurements from periodic sampling and the increasing use of continuous in situ measurements both increase the need for proper modeling of weekly and diurnal cycles.

[37] Using the Vulcan v2.0 emission data product for the United States and emission sector information from EDGAR v4.2 for the 20 highest-emitting countries, along with information on weekly cycles from satellite observations of NO<sub>2</sub> and other sources, we derive scale factor maps that can be applied to the global inventories to represent weekly and diurnal variability. We refer to this new product as TIMES (Temporal Improvements for Modeling Emissions by Scaling). Because Canada's *NIR* [2011] provides information on the differences between the contributions from different sectors among the Canadian provinces, we use provincial information for Canada rather than EDGAR national sector contributions.

[38] Surface atmospheric impacts of the combined diurnal and weekly scaling were determined in GEOS-Chem and shown to be negligible for locations very far from emission sources but in the range of ~1.5-8 ppm over large urban areas (Los Angeles, New York City, Berlin, Tehran, and Beijing). These numbers should be contrasted with the accuracy (~0.1 ppm) and precision (~0.2 ppm) of flask [*Conway et al.*, 1994] and continuous in situ [*Worthy et al.*, 2003] CO<sub>2</sub> measurements. Impacts of the combined temporal scaling on XCO<sub>2</sub> were found to reach as high as 0.1-0.5 ppm over the same major urban areas, with a larger proportion resulting from the weekly variability than with surface CO<sub>2</sub>, which will be relevant for the assimilation of XCO<sub>2</sub> satellite observations.

[39] We also derive scale factors to account for the large per capita differences in CO<sub>2</sub> emissions between Canadian provinces. The NIR reported high per capita emissions for Alberta (13.68 Mg C person<sup>-1</sup> yr<sup>-1</sup>), related to the dependence on coal for electricity generation as well as the fossil fuel extraction and refining industries, and for Saskatchewan  $(11.94 \text{ Mg C person}^{-1} \text{ yr}^{-1})$ , for the same reasons, along with high off-road mobile emissions, presumably from farming. Low per capita emissions are found for some other provinces, including Quebec  $(2.19 \text{ Mg} \text{ C person}^{-1} \text{ yr}^{-1})$  and Manitoba  $(2.57 \text{ Mg C person}^{-1} \text{ yr}^{-1})$ , because of the high proportion of hydroelectric generation, and Ontario  $(2.92 \text{ Mg C person}^{-1} \text{ yr}^{-1})$ , because of the high proportion of non-CO<sub>2</sub>-emitting electricity generation methods including nuclear, hydroelectric, and renewable energy, although differences in per capita total energy use are also a factor. Quantification of the impacts of these spatial scale factors for Canada will be investigated in future work.

[40] The resulting products of this work are a set of 24 diurnal and seven weekly global  $0.25^{\circ} \times 0.25^{\circ}$  gridded temporal scale factor maps that can be applied to common global fossil fuel CO<sub>2</sub> emission inventories to represent weekly and diurnal variability and a set of  $1^{\circ} \times 1^{\circ}$  spatial scale factor maps to adjust CDIAC and ODIAC to account for variability in per capita emissions within Canada. Both sets of scale factors (along with an additional set of weekly factors with no scaling for China and its proxy countries, based on the NO<sub>2</sub> analysis of *Beirle et al.* [2003]) are publicly available from CDIAC website. Implementation of these temporal and spatial scale factors should contribute to a reduction in systematic errors related to CO<sub>2</sub> emissions from fossil fuel combustion and cement manufacture in the current generation of inverse modeling and data assimilation systems, which typically impose fossil fuel emissions and optimize fluxes from the terrestrial biosphere and oceans using atmospheric CO<sub>2</sub> point measurements. Scale factors such as these will be crucial for the more sophisticated systems being developed for assimilation of satellite CO<sub>2</sub> observations, intended for estimation of terrestrial and ocean fluxes at higher spatial and temporal scales. Future systems could also be used for deriving optimized estimates of CO2 from fossil fuel emissions, in which these scale factors could be applied to yield improved temporal and spatial distributions in a priori estimates, which should lead to more accurate CO<sub>2</sub> emission quantification.

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