

Monthly carbon emissions from natural-gas flaring and cement manufacture in the United States

By T. J. BLASING^{1*} and KIMBERLY HAND², ¹*Environmental Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831-6335, USA;* ²*School of Chemical and Biochemical Engineering, Georgia Institute of Technology, Atlanta, GA 30332, USA*

(Manuscript received 14 December 2005; in final form 27 June 2006)

ABSTRACT

Annual data on carbon emissions from fossil-fuel combustion and cement manufacture have been used in studies of the carbon cycle for the last few decades. However, annual data do not specify carbon emissions on the seasonal timescales relevant to biospheric uptake and other processes affecting the carbon cycle. Estimates of monthly emissions from fossil-fuel consumption in the United States (US) have shown that an increasing percentage of the annual emissions are occurring during the growing season; however, carbon emitted from flaring natural gas at well sites was not accounted for in those emissions estimates, nor was carbon emitted during cement manufacture. Here we show that emissions from flaring, which amount to around 0.1 % of all fossil-fuel carbon emissions in the US, have no clear and persistent annual pattern that can be detected in the data. In contrast, carbon emissions from cement manufacture, which add about 0.7% to carbon emissions from fossil fuels in the US, have a clear and persistent annual pattern including low values in late winter and early spring. In this paper, we provide a few remarks on carbon emissions from natural-gas flaring before presenting monthly emissions estimates. We then focus on the methodology for calculating carbon emissions from cement manufacture before presenting and discussing the monthly emissions estimates.

1. Introduction and overview

Marland et al. (2005) have provided estimates of annual emissions of carbon, as carbon dioxide (CO₂), from fossil-fuel combustion (including natural-gas flaring) and from cement manufacture, for all countries of the world. These estimates are useful for tracking emissions at the global scale and for each country, for measuring progress towards meeting emissions-reduction goals, and for providing large-scale constraints on the spatial distribution of carbon emissions. However, annual totals provide no information on the seasonal distribution of emissions, which is an important consideration in carbon-cycle modelling. Temperature, daylength, available water and photosynthesis are examples of seasonally varying influences on biospheric uptake of atmospheric CO₂. During the growing season, elevated CO₂ concentrations near large emissions sources can lead to increased biospheric uptake, as measured by net primary productivity. This could represent long-term carbon sequestration in the terrestrial biosphere if the productivity is not manifest in rapid-turnover products such as leaves and fine roots (IPCC 2001a; Norby et al., 2002). Because the processes involved occur at small spatial

and temporal scales, understanding the mechanisms involved requires finer spatial and temporal resolution of fossil-fuel carbon emissions than annual values on a nation-by-nation basis. Blasing et al. (2005) have begun to increase the temporal resolution by providing estimates of monthly fossil-fuel carbon emissions for the United States (US); their results indicate that over the last 25 yr an increasing fraction of the annual emissions has occurred during the growing season.

This report is intended to supplement the monthly fossil-fuel carbon emissions presented by Blasing et al. (2005), which did not include carbon from gas flaring. That paper did include emissions from fossil fuels used in cement manufacture but did not address the carbon dioxide released from the heating of carbonates in the raw materials during cement production (calcination). Here, we present estimates of monthly carbon emissions from flaring of natural gas at well sites, and from calcination during cement manufacture in the US.

Fossil-fuel consumption in the US accounts for over 1500 teragrams (Tg; 1 Tg = 10¹² g) of carbon emissions per year, and flaring currently increases this total by only 1–2 Tg. Calcining during cement manufacture currently accounts for about 10–12 Tg annually. Although CO₂ emitted from these sources combined is only around 1% of US fossil-fuel emissions, such small amounts are nontrivial when setting emissions goals or tracking progress toward meeting them. Moreover, as noted above,

*Corresponding author.
e-mail: blasingtj@ornl.gov
DOI: 10.1111/j.1600-0889.2006.00195.x

elevated CO₂ concentrations near large emissions sources can affect the local cycling of carbon, including its removal from the atmosphere. In the following material we first discuss methods and assumptions involved in estimating carbon emissions from flaring of natural gas, and present our estimates of monthly values for the US (Section 2). We then provide a discussion of the methodology for estimating carbon emissions from cement manufacture (Section 3) and present our monthly estimates (Section 4). Conclusions are given in Section 5.

2. Natural gas flaring

Natural gas typically occurs in association with oil deposits and oil wells. At well sites, this gas is sometimes reinjected into the ground for storage if later recovery appears economically advantageous, and/or to increase well pressure as an aid to pumping. Typically, however, it is released at the well sites. In the US this is usually done by flaring (combustion in an open flame) rather than by venting (releasing uncombusted natural gas). Controlled flaring prevents the gas from remaining near the site in sufficiently high concentrations that it could potentially ignite, leading to uncontrolled burning. It also oxidizes chemicals that may lead to asphyxiation if allowed to remain unoxidized in the high concentrations that occur near their sources. On much larger spatial scales, the flaring of natural gas has implications for global change. The Intergovernmental Panel on Climate Change (IPCC) lists methane (CH₄), the main component of natural gas, as about 62 times more effective than CO₂ as a greenhouse gas on a 20 yr timescale (IPCC, 2001b). Although CH₄ is eventually destroyed as the carbon goes into CO₂, the atmospheric lifetime of CH₄ is around 10 yr (depending on whether feedbacks are considered, IPCC, 2001b); flaring transforms the methane immediately.

Only the total amounts of natural gas released at the well sites are reported. Fractions combusted to CO₂, released as uncombusted CH₄, and released as soot have not been well characterized in the literature. These fractions are important because CO₂, CH₄, and soot have substantially different impacts on the environment. As noted above, CH₄ is a more effective greenhouse gas than CO₂ and soot particles can lead to atmospheric cooling rather than warming. Barns and Edmonds (1990) gave an estimate of 13.3% of the total flared/vented gas that was ‘actually vented’ and assumed that around 92% of the carbon in the remaining gas was oxidized to CO₂ (or to carbon monoxide, CO, which is converted directly to CO₂ by the hydroxyl radical) given the flaring technologies at the time. Additional regulations against flaring and improved technologies have probably increased the percentage of carbon converted to CO₂ in flared/vented gas over the last 15 yr, but little light has been shed on this subject over that period, and considerable uncertainty and resulting controversy still remain.

The US Environmental Protection Agency (EPA, 2006) distinguishes between ‘onshore’ and ‘offshore’ flaring/venting, and

gives rather low percentages (e.g. 26% in 2004) for the amount that is flared offshore. Their estimated offshore amounts of carbon emitted as CO₂ would add around 3% to our total carbon emissions from flaring, which only include ‘onshore’ activities. Of course, these percentages would be vastly different when accounting for carbon emitted as CH₄.

Incomplete or inconsistent reporting may result in some artificial variability being added to the data, or to some actual variability being smoothed out, or both. Increases in reporting frequency tend to reduce the need for estimation, including smoothing; decreases in reporting frequency have the opposite effect. Additionally, some consistent over-reporting identified by EIA (2001) has only been corrected back through 1998.

The percentage of input carbon that is oxidized by a flare is also controversial, and terms like ‘flaring efficiency’ are often used without a complete definition. EPA (1995) stated that the combustion efficiency for a ‘properly operated’ flare is at least 98%. A ‘flaring efficiency’ value of 98% appeared in a paper cited by IPCC (2000, page 2.87, footnote e). It is not clear what factors were included in the determination of that ‘flaring efficiency’ but the 98% figure is higher than the 92% figure assumed by Barns and Edmonds (1990) which allowed for incomplete combustion and periods of flare extinction.

In view of the uncertainties involved in determining the amount of reported flared/vented gas that is actually flared, and the lack of precision for the large percentage of carbon that is oxidized in a flare, we take an approach used by EPA (2006), and present full-combustion values for all reported flared/vented (onshore) natural gas. Full-combustion values are the amounts of emitted CO₂ if all flared/vented carbon is flared, all flared carbon actually enters a flame and is immediately converted to CO₂, and volumes are reported precisely (there are no missing values and no ‘over-reporting’ has occurred due to such things as inadvertent double counting).

This approach is simply a convenient default methodology, which is used because of the large number of unknown factors involved. Our values do not include offshore flaring in the Gulf of Mexico. One may multiply our (‘onshore’) estimates by a factor representing some combination of influences not included in our approach, and such a factor does not necessarily have to be less than 1.0 if it can be argued that emissions have been systematically under-reported. Our objective here was to examine the emissions estimates for any clear and repetitive annual pattern of higher values during some calendar months and lower values during others.

Total monthly values of ‘vented and flared’ natural gas in the US for years 1980–2004 were obtained from the Energy Information Administration (EIA) of the US Department of Energy. However, monthly values through 1997 have not been corrected for an over-reporting problem noted by EIA (2001), so we only report results for 1998–2004 in this paper. The volumes are given in cubic feet (ft³), where 1 ft³ = 0.0283 m³. Recent values (in 10⁹ ft³) are published monthly, by EIA, in Table 4.2 of *Monthly*

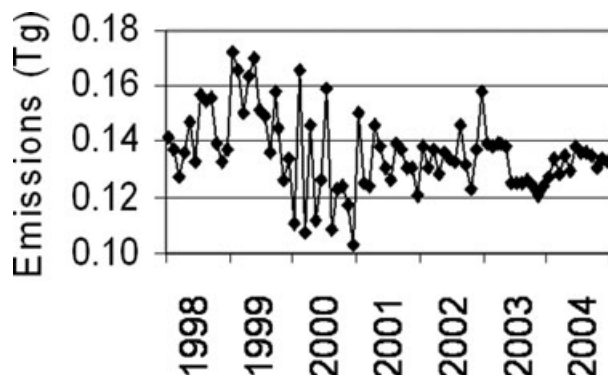


Fig. 1. Estimates of monthly US carbon emissions from gas flaring, scaled to an equal number of days in each month.

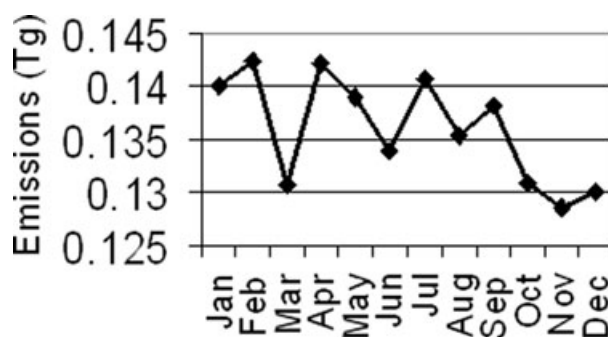


Fig. 2. Seven-year (1998–2004) average US carbon emissions from gas flaring for each calendar month, scaled to an equal number of days in each month.

Energy Review. These volumes of natural gas were converted to estimates of carbon emissions using the formula:

$$C = G \bullet 1106 \bullet 14.92,$$

where C is the carbon (Tg) emitted as CO₂; G is the amount of natural gas flared (in 10¹⁵ ft³); 1106 is the thermal conversion factor, in Btu/ft³, where 1 Btu = 1055 joules, and 14.92 is the carbon coefficient for 100% oxidation to CO₂, in Tg-C/10¹⁵ Btu. The thermal conversion factor (1106) and the carbon coefficient (14.92) were obtained from EIA (2005a).

Monthly estimates of US carbon emissions from flaring of natural gas for January 1998 to December 2004 are shown in Fig. 1. These data were scaled so each calendar month represents an equal number of days. There is an interesting decrease in variance during the later years (2003–2004), which could result from any of a number of factors, including an increased need for smoothing if the reporting frequency of monthly values decreased.

Seven-year 1998–2004 averages of the estimated emissions, for each calendar month, are shown in Fig. 2. These data were also scaled to an equal number of days in each month. The averages indicate a tendency for lower values during 3 successive months at the end of the calendar year, when increasing demand

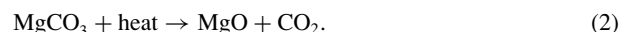
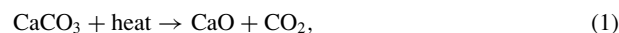
may tend to make it more profitable to market the gas rather than to flare it. However, 7 yr averages calculated from such highly variable data may well be artefacts, even when three successive months stand out clearly as being below the annual mean. Because much of the variance of the time-series (Fig. 1) is in the lower frequencies, the variance clearly changes with time and only seven data points are included in each monthly average in Fig. 2, the value of significance testing is questionable. We will simply state that no clear and persistent annual pattern is evident in the data shown in Fig. 1, and uncertainties involved in estimating the amount of natural gas flared each month may tend to obscure any such pattern that might exist.

3. Cement manufacture

Cement is made from limestone and other carbonates that have been crushed and burned. The generic technique was known to the Romans, who mixed the crushed, burned limestone with volcanic ashes. The volcanic ash came from Pozzouli; hence, the name *pozzolan* is still used for any substance containing silicon that develops cementing properties when mixed with lime and water.

Today, limestone, clay, sand, fly ash, slag and other substances are mixed and heated in a kiln to temperatures of around 1500 °C to form a substance called clinker. After the clinker has cooled, it is finely ground and mixed with a small amount of gypsum (about 5% of the mass of the clinker) to prevent the cement from hardening too rapidly. The final product was patented by Joseph Aspdin in 1824. He called it *Portland cement* because it resembled stone quarried from the Isle of Portland, off the south coast of England.

The mixture of materials that goes into clinker includes, among other things, calcium carbonate (CaCO₃) and magnesium carbonate (MgCO₃). During the heating process, carbon dioxide (CO₂) is produced as the following reactions take place:



These equations describe the sources of CO₂ released from carbonates in the raw materials during the process known as calcination. Other carbonates or bicarbonates (e.g. sodium bicarbonate) may be present in the raw materials, but only in trace amounts. The CO₂ originating from CaCO₃ accounts for almost all the CO₂ emitted during calcination. The MgCO₃ may contribute a small amount, and there is still some controversy about whether or not to include it in emissions inventories. The calculation of the CO₂ emitted during calcination is based on the amount of the resulting oxides (CaO or MgO) in the clinker and the assumption that all of these oxides originated from carbonates in the raw materials. Because small amounts of these oxides are likely to be present in the raw materials before calcination, the

assumption that they all originated from CO₂ emissions during calcination leads to a high bias in the resulting CO₂ emissions estimates. That bias would be augmented, rather than compensated, by including CO₂ from MgCO₃. For that reason, and because amounts of MgCO₃ in the raw materials are small and not well characterized, we follow the convention of IPCC (1997, 2000), and do not include the MgCO₃ source in our calculations. However, we provide the necessary stoichiometric calculations below for those who wish to include it.

Some fine-grained material produced in the kiln is left behind as cement kiln dust (CKD), and is not reported as clinker produced. The CKD can amount to several per cent of the clinker produced, but in modern plants it is largely recovered using electrostatic precipitators and/or filtration, so that lost, or fugitive, CKD is only around 2% of the reported clinker produced. For the calculation of CO₂ emissions from clinker production, IPCC good practice guidance (IPCC, 2000) recommends adding 2% to the reported clinker-production value. This recommendation is followed by EPA (2006).

According to IPCC (1997) and EPA (2006), the average CaO content of clinker is 64.6%. EPA (1995) gave a figure of around 63.5% for *cement*, which is about 95% clinker. That same figure (63.5% of *cement*) is given by Griffin (1987) in a study cited by Andres et al. (2000), and used by Marland et al. (2005). Distinctions between cement and clinker are important because only cement data are available for many countries, but the use of clinker data, if available, yields more precise values because the amount of added gypsum does not have to be approximated by an estimated average. We used the 64.6 percentage for clinker, rather than the percentage given earlier for *cement*, so assuming that cement is 95% clinker, our results correspond to a value of $64.6 \times 0.95 = 61.4\%$ as the percentage of *cement* that is CaO. This is lower than the previously used value (63.5%) for *cement*. The amount of added gypsum would have to be around 1.7% for the 63.5% figure for *cement* to agree with the 64.6 in clinker.

We do not include imported clinker in our calculations because the associated CO₂ emissions occurred in other countries, and emissions are conventionally assigned to the country where they occur. This convention is useful for purposes of setting and maintaining international protocols to reduce emissions, and for global carbon-cycle models in which locations of the actual CO₂ sources are important. However, some political entities could reduce their CO₂ emissions by transferring emissions-intensive activities outside their boundaries while continuing to purchase the products of those activities. Such practice would transfer the emissions burden but would not necessarily reduce the global source term.

Because Marland et al. (2005) use the higher percentage (63.5) for the amount of CaO in *cement*, they obtain higher values than will result from this study. Additionally, Marland et al. (2005) use figures for cement production from Table 23 of *Minerals Yearbook* published annually by the United States Geological Survey (USGS). The advantage of using these figures is that they

provide uniform estimates of cement manufacture for over 150 countries. However, for the US, the figure includes cement made from imported clinker. Additionally, the *Minerals Yearbook* data combine Portland and masonry cement, whereas we treat those types of cement separately, below. Marland et al. (2005) do subtract cement made in Puerto Rico from the US total; otherwise their result would be even higher. The imported clinker adds a small percentage, which varies from year to year, to the total (*Minerals Yearbook*, Table 1). Over the 5 yr (1998–2002) common to this study and Marland et al. (2005), the average percentage added by imported clinker was 3.9. The inclusion of masonry cement according to the methodology of Marland et al. (2005) added another 5.2%, but the production of masonry cement contributes only negligibly to atmospheric carbon emissions, as shown below. Thus, the totals given by Marland et al. (2005) would be expected to be about $63.5/61.4 \times 1.039 \times 1.052 (=1.131)$ times the results obtained by our methodology if only CO₂ emissions from reported clinker production were included, and about $1.13/1.020 (=1.109)$ times the results obtained by our methodology if 2% is added to our clinker totals to account for CKD. Because the last decimal place was included only to prevent the accumulation of round-off error, we conclude that the results of Marland et al. (2005) are expected to be around 10–11 percent higher than those calculated in this study. As noted by Blasing et al. (2005), the figures presented by Marland et al. (2005) are appropriate to comparing one country to another, because data for all countries are derived using the same assumptions. However, more detailed data allow more precise estimates for the US, so we believe our results are a more precise indicator of carbon emissions from cement manufacture in the US.

In the following material we present our methodology in some detail so that differences in carbon emissions estimates arrived at by various accounting strategies may be more easily reconciled.

3.1. Overview of methodology

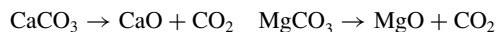
Our basic data is clinker production, taken from Table 4a of *Mineral Industry Surveys*, published monthly by the USGS. We subtracted the production from Puerto Rico to arrive at a monthly total clinker production from the US.

In our calculations below, we have chosen to neglect CO₂ from masonry cement and from magnesium carbonate. We do not recommend trying to incorporate masonry cement because: (1) for many accounting purposes, that CO₂ is already accounted for under lime manufacture and (2) in any case, as we show in Section 3.3, below, it is typically too small an amount to affect the final result to the third decimal place. For those who wish to include it, the calculations are given below.

3.2. Stoichiometry

The chemical reactions leading to CO₂ release during clinker formation are given below, followed in the next line by equations

giving the molecular weights of the compounds involved, from the periodic table of the elements.



$$100.09 = 56.08 + 44.01 \quad 84.32 = 40.31 + 44.01.$$

These equations show that 44.01 g of CO₂ are produced for every 56.08 g of CaO, and for every 40.31 g of MgO, in the clinker and CKD.

Using 64.6 as the percentage of clinker that is CaO (IPCC, 1997, EPA 2006) and x as the percentage of clinker that is MgO, we can estimate the amount of CO₂ released from each gram of clinker produced.

From calcium carbonate:

$$0.646 \times (44.01/56.08)$$

= 0.507 grams of CO₂ per gram of clinker produced.

From magnesium carbonate:

$$x/100 \times (44.01/40.31)$$

= 0.011 x grams of CO₂ per gram of clinker produced.

Multiplying by 12.01/44.01 (= 0.273) to convert CO₂ to carbon, we obtain:

$$0.507 \times 0.273 = 0.138 \text{ grams of carbon per gram of}$$

reported clinker from calcium carbonate, and

$$0.011x \times 0.273 = 0.003x \text{ grams of carbon per gram of}$$

reported clinker from magnesium carbonate.

Combining these results to estimate the carbon emitted (C) from CaCO₃ and MgCO₃ in the raw materials, we obtain:

$$C = (0.138 + 0.003x)(\text{mass of clinker}). \quad (3)$$

Accounting for CKD, we multiply carbon emissions per gram of clinker 0.138 + 0.003 x by 1.02 to obtain:

$$C = (0.138 + 0.003x)(1.02)(\text{mass of clinker}),$$

or, rounding to the third significant figure,

$$C = (0.141 + 0.003x)(\text{mass of clinker}). \quad (4)$$

Thus, if we neglect MgCO₃ in the raw materials, x becomes zero in equation 4, and we can estimate the carbon emissions from cement manufacture as:

$$C = 0.141(\text{mass of clinker}).$$

If, for example, we assume that 2% of the clinker is MgO originating from MgCO₃ in the raw materials, then the carbon emissions estimate becomes (0.141 + 0.006) = 0.147 times the mass of the clinker produced.

3.3. Contribution of masonry cement

Because additional lime (CaO) is used in masonry cement, CO₂ emitted during production of this lime may be included in the emissions totals. This may result in double counting if the same CO₂ is included in a separate 'lime production' category. In any case, inclusion of this CO₂ adds only a very small percentage to the amount obtained by excluding it, as shown below.

According to the year-end summary in the monthly publication, *Mineral Industry Surveys*, in 2004 (the latest year for which annual data were available), 5.114 Tg of masonry cement were produced in the US (none was produced in Puerto Rico). This includes some cement made from imported clinker. According to EPA (2006), 2.86% of this amount was additional lime not counted in clinker. Assuming this additional lime is all CaO then, from the stoichiometry, (12.01/56.08) = 0.214 g of carbon (as CO₂) are produced for each gram of CaO. Multiplying 0.0286 $\times$ 0.214 = 0.00612, and multiplying 0.00612 by the amount of masonry cement produced (5.114 Tg) gives 0.031 Tg as the amount of carbon emitted as CO₂.

Another way to estimate this emitted carbon is to use the formula given by IPCC (1997). In this formulation, the amount of carbon, C , as CO₂, attributable to lime used in masonry cement is:

$$C = [1 - 1/(1 + b)](c)(0.214)(\text{amount of masonry cement produced})$$

where C is the mass of carbon produced; b is the decimal fraction of mass added to masonry cement by non-plasticizer additives such as lime, slag, and shale; and c is the decimal fraction of mass of non-plasticizer additives that is lime. Central values for b and c are, respectively, 0.04 and 0.7 (IPCC, 1997). Substituting these values, the amount of carbon, as CO₂, from the production of the lime used in masonry cement is:

$$[1 - 1/(1 + 0.04)](0.7)(0.214)(5.114\text{Tg}) = 0.029\text{Tg},$$

which compares well with 0.031 Tg, derived above.

In addition, EIA (2005b) gives 0.1 Tg as the amount of CO₂ emitted from US production of masonry cement for recent years; this translates to around 0.03 Tg of carbon. Therefore, we use 0.03 Tg as the carbon emissions from production of additional lime added to masonry cement. As will be shown later, this is less than 0.3% of the total national carbon emissions from cement manufacture in 2004. Also, it is often reported under a separate category for lime manufacture (e.g. IPCC Source Category 2A2); it should not be double-counted in such cases.

4. Results for cement manufacture

Estimates of US carbon emissions from cement manufacture, for each month from 1998 to 2004, are shown in Fig. 3. A clear and persistent annual pattern is evident. These data were

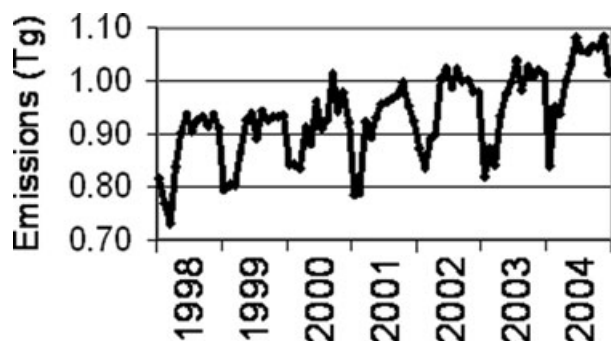


Fig. 3. Estimates of monthly US carbon emissions from cement manufacture, scaled to an equal number of days in each month.

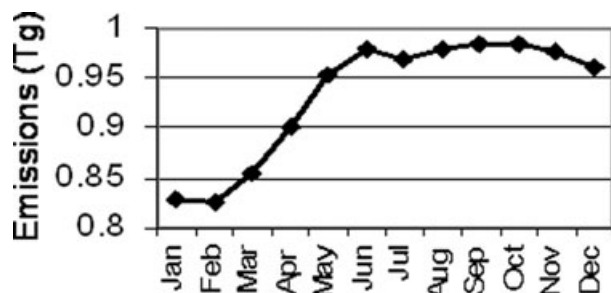


Fig. 4. Seven-year (1998–2004) average US carbon emissions from cement manufacture for each calendar month, scaled to an equal number of days in each month.

scaled so each calendar month represents an equal number of days; therefore the tendency for low values for February is not an artefact of the data processing. Total carbon emissions from cement manufacture in the US during 2004 were estimated as 12.206 Tg.

Marland et al. (2005) give carbon emissions from cement manufacture in the US for years 1928 to 2002. For the 5 yr (1998–2002) of their data in common with ours, their results average 8.4% higher. From the introductory material in Section 3, this percentage would be expected to be around 10.9%. Differences between *Minerals Yearbook* and the annual totals given in *Mineral Industry Surveys* (our source) regarding the amount of clinker production reported account for only 0.2% percent of the difference, suggesting that an unknown factor, which accounts for around 2–2.5% of one or the other set of values, is involved.

Figure 4 shows the 7 yr average carbon emissions estimates for each calendar month. Monthly averages are lowest during January to March; April is a transition month and the monthly averages are relatively equal from May to December. Lowest values during the late winter and spring seasons may be related to unfavourable weather conditions for outdoor construction activities. The high values extending into the beginning of winter may be due to stockpiling or other factors.

5. Conclusions

We have estimated monthly US carbon emissions from gas flaring and from chemical processes involved in cement production. This work supplements our earlier calculations of monthly US carbon emissions from fossil fuel combustion (Blasing et al., 2005), which did not include gas flaring but did include emissions from fossil-fuel combustion during cement production.

No clear repetitive annual pattern of monthly carbon emissions from gas flaring appears in our results. Observed multi-year trends may result from economic considerations of supply and demand, but further analysis is necessary to explain these variations. Reporting problems may be involved, but their relationship to the longer-term variations is not clear. Incomplete or inconsistent reporting and failure to distinguish between flaring and venting can either add variance to the data that is unrelated to flaring or smooth out variance that is due to flaring. Consistent under-reporting is possible, and inadvertent over reporting has been noted above. Such problems may be at least partly responsible for our failure to identify any annually repetitive pattern in the monthly data.

Emissions from cement production show a clear repetitive annual pattern, with lowest values in late winter and early spring. Suitability of weather conditions for the initiation of outdoor construction activity may be involved. Such repetitive annual sequences are important in the larger framework of carbon-cycle modelling; for example, the fraction of emitted CO_2 sequestered by the terrestrial biosphere is likely to differ from one season to the next.

Whether or not an annual pattern of high values in some seasons and low values in others is present, monthly totals can serve as constraints on emissions estimates integrated over finer temporal scales. Such estimates can be based on daily and weekly patterns of traffic, gas usage in residential and commercial buildings etc. Such fine-scale carbon emissions estimates are important because carbon-cycling processes also depend on factors exhibiting daily cycles, such as temperature, and, during the growing season, on photosynthetic activity. Because these factors are likely to be most important near large emissions sources, where resulting CO_2 concentrations are highest, increasing the spatial resolution of carbon emissions inventories is also important.

Data presented here, and updates thereof, are available from the Carbon Dioxide Information Analysis Center at Oak Ridge National Laboratory. Because these data may be used for many purposes, some of which may involve only one calendar month, these data have not been adjusted for the varying number of days from one calendar month to the next.

6. Acknowledgements

This work was supported by the Biological and Environmental Program of the Office of Science, US Department of Energy. Oak

Ridge National Laboratory is managed by UT-Battelle, LLC, under Contract No. DE-AC05-00OR22725 for the US Department of Energy. We thank Tom Boden and Greg Zimmerman, Environmental Sciences Division, Oak Ridge National Laboratory, for internal reviews of this manuscript. We also thank Lisa Hanle of the U.S. Environmental Protection Agency, and one anonymous reviewer for their suggestions which substantially improved the manuscript. Data, along with helpful suggestions for interpreting it, were provided by Roy Kass, Energy Information Administration, US Department of Energy (flaring/venting data), and by Hendrik van Oss and Robert Callaghan, US Geological Survey (clinker data). Louise Clemens, a student at Oak Ridge High School, Oak Ridge, TN was involved in early stages of this work.

References

- Andres, R. J., Marland, G., Boden, T. and Bischof, S. 2000. Carbon dioxide emissions from fossil fuel consumption and cement manufacture, 1751–1991, and an estimate of their isotopic composition and latitudinal distribution. In: *The Carbon Cycle* (eds T. M. L. Wigley and D. S. Schimel). Cambridge University Press, Cambridge, pp. 53–62.
- Barns, D. W. and Edmonds, J. A. 1990. *An Evaluation of the Relationship Between the Production and Use of Energy and Atmospheric Methane Emissions*. U.S. Department of Energy, DOE/NBB-0088P, TR047, p. 3.7.
- Blasing, T. J., Broniak, C. T. and Marland, G. 2005. The annual cycle of fossil-fuel carbon dioxide emissions in the United States. *Tellus* **57B**, 107–115.
- EIA (Energy Information Administration, US Department of Energy). 2001. *Natural Gas Annual 2000*, DOE/EIA-0131(00). US Department of Energy, Washington, DC, p. 3.
- EIA (Energy Information Administration, US Department of Energy). 2005a. *Documentation for Emissions of Greenhouse Gases in the United States 2003*. DOE/EIA-0638 (2003). US Department of Energy, Washington, DC, p. 6 for combustion efficiencies; p. 35 for thermal and carbon coefficients used in flaring.
- EIA (Energy Information Administration, US Department of Energy). 2005b. *Emissions of Greenhouse Gases in the United States 2004*. DOE/EIA-0573(2004). US Department of Energy, Washington, DC, p. 34.
- EIA (Energy Information Administration, US Department of Energy). Various dates. *Monthly Energy Review*. Available online at: <http://www.eia.doe.gov/emeu/mer/contents.html>.
- EPA (United States Environmental Protection Agency). 1995. *Compilation of Air Pollutant Emission Factors, Volume 1. Stationary Point and Area Sources*. AP-42, 5th Edition, pp. 11.6–6 for cement manufacture; pp. 13.5–3 for gas flaring.
- EPA (United States Environmental Protection Agency). 2006. *Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990–2004* (Section 3.10 for flaring; Section 4.2 for cement manufacture; Table A-27 for combustion efficiencies).
- Griffin, R. C. 1987. *CO₂ Release from Cement Production 1950–1985*. Institute for Energy Analysis, Oak Ridge Associated Universities, Oak Ridge, TN.
- IPCC (Intergovernmental Panel on Climate Change). 1997. *Greenhouse Gas Inventory Reference Manual, Revised 1996 IPCC Guidelines for National Greenhouse Gas Inventories*, **3**, p 1.116 for flaring; pp. 2.6–2.7 for cement manufacture.
- IPCC (Intergovernmental Panel on Climate Change). 2000. *Good Practice Guidance and Uncertainty Management in National Greenhouse Gas Inventories* (Section 2.7 for flaring and Section 3.1.1, especially footnotes 1 and 2 on p 3.12, for cement production).
- IPCC (Intergovernmental Panel on Climate Change). 2001a. *Climate Change 2001: Impacts, Adaptation and Vulnerability*. p. 294.
- IPCC (Intergovernmental Panel on Climate Change). 2001b. *Climate Change 2001: The Scientific Basis* (Table 6.7).
- Marland, G., Boden, T. A. and Andres, R. J. 2005. Global, Regional, and National CO₂ Emissions. In: *TRENDS: A Compendium of Data on Global Change*. Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, U.S. Department of Energy, Oak Ridge, TN.
- Norby, R. J., Hanson, P. J., O'Neill, E. G., Tschaplinski, T. J., Weltzin, J. F. and co-authors. 2002. Net primary productivity of a CO₂-enriched deciduous forest and the implications for carbon storage. *Ecological Applications* **12**, 1261–1266.
- United States Geological Survey (USGS). *Minerals Yearbook Vol. 1, Metals and Minerals*. Issued annually by USGS, U.S. Department of the Interior, 988 National Center, Reston VA, 20192, USA. <http://minerals.usgs.gov/minerals/pubs/commodity/cement/index.html#myb>
- United States Geological Survey (USGS). 1998–2005. *Mineral Industry Surveys*, monthly, USGS, U.S. Department of the Interior, 988 National Center, Reston VA, 20192, USA. <http://minerals.usgs.gov/minerals/pubs/commodity/cement/index.html#mis>