

# UC Irvine

## Faculty Publications

### Title

An atmospheric perspective on North American carbon dioxide exchange: CarbonTracker

### Permalink

<https://escholarship.org/uc/item/7jw635bt>

### Journal

Proceedings of the National Academy of Sciences, 104(48)

### ISSN

0027-8424 1091-6490

### Authors

Peters, W.  
Jacobson, A. R.  
Sweeney, C.  
et al.

### Publication Date

2007-11-01

### DOI

10.1073/pnas.0708986104

### Copyright Information

This work is made available under the terms of a Creative Commons Attribution License, available at <https://creativecommons.org/licenses/by/4.0/>

Peer reviewed

# An atmospheric perspective on North American carbon dioxide exchange: CarbonTracker

Wouter Peters<sup>\*†‡</sup>, Andrew R. Jacobson<sup>\*†</sup>, Colm Sweeney<sup>\*†</sup>, Arlyn E. Andrews<sup>\*</sup>, Thomas J. Conway<sup>\*</sup>, Kenneth Masarie<sup>\*</sup>, John B. Miller<sup>\*†</sup>, Lori M. P. Bruhwiler<sup>\*</sup>, Gabrielle Pétron<sup>\*†</sup>, Adam I. Hirsch<sup>\*†</sup>, Douglas E. J. Worthy<sup>§</sup>, Guido R. van der Werf<sup>¶</sup>, James T. Randerson<sup>||</sup>, Paul O. Wennberg<sup>\*\*</sup>, Maarten C. Krol<sup>††</sup>, and Pieter P. Tans<sup>\*</sup>

<sup>\*</sup>National Oceanic and Atmospheric Administration Earth System Research Laboratory, 325 Broadway R/GMD1, Boulder, CO 80305; <sup>†</sup>Cooperative Institute for Research in Environmental Sciences, UCB 216, Boulder, CO 80303; <sup>§</sup>Environment Canada, 4905 Dufferin Street, Downsview, ON, Canada M3H 5T4; <sup>¶</sup>Faculty of Earth and Life Sciences, Vrije Universiteit, 1081 HV, Amsterdam, The Netherlands; <sup>||</sup>Department of Earth System Science, University of California, Irvine, CA 92697; <sup>\*\*</sup>Division of Engineering and Applied Science and Division of Geological and Planetary Sciences, California Institute of Technology, Pasadena, CA 91125; and <sup>††</sup>Department of Meteorology and Air Quality, Wageningen University and Research Center, 6708 PB, Wageningen, The Netherlands

Communicated by A. R. Ravishankara, National Oceanic and Atmospheric Administration, Boulder, CO, September 27, 2007 (received for review May 23, 2007)

We present an estimate of net CO<sub>2</sub> exchange between the terrestrial biosphere and the atmosphere across North America for every week in the period 2000 through 2005. This estimate is derived from a set of 28,000 CO<sub>2</sub> mole fraction observations in the global atmosphere that are fed into a state-of-the-art data assimilation system for CO<sub>2</sub> called CarbonTracker. By design, the surface fluxes produced in CarbonTracker are consistent with the recent history of CO<sub>2</sub> in the atmosphere and provide constraints on the net carbon flux independent from national inventories derived from accounting efforts. We find the North American terrestrial biosphere to have absorbed  $-0.65$  PgC/yr (1 petagram =  $10^{15}$  g; negative signs are used for carbon sinks) averaged over the period studied, partly offsetting the estimated  $1.85$  PgC/yr release by fossil fuel burning and cement manufacturing. Uncertainty on this estimate is derived from a set of sensitivity experiments and places the sink within a range of  $-0.4$  to  $-1.0$  PgC/yr. The estimated sink is located mainly in the deciduous forests along the East Coast (32%) and the boreal coniferous forests (22%). Terrestrial uptake fell to  $-0.32$  PgC/yr during the large-scale drought of 2002, suggesting sensitivity of the contemporary carbon sinks to climate extremes. CarbonTracker results are in excellent agreement with a wide collection of carbon inventories that form the basis of the first North American State of the Carbon Cycle Report (SOCCR), to be released in 2007. All CarbonTracker results are freely available at <http://carbontracker.noaa.gov>.

carbon cycle | greenhouse gases | data assimilation | biogeochemistry | atmospheric composition

Projections of future CO<sub>2</sub> levels in the atmosphere and the associated climate forcing, as well as our ability to control CO<sub>2</sub> levels, depend substantially on our scientific understanding of the natural carbon cycle. Its current capacity to absorb close to half of the carbon released from fossil fuel burning is not guaranteed to grow along with rapidly rising man-made emissions or to even continue at its present-day magnitude. Moreover, natural emissions themselves might increase as a result of already observable rapid warming in parts of the Arctic (1), where large carbon reservoirs are buried beneath the permafrost. Major national and international programs to study the carbon cycle are therefore underway.

The National Oceanic and Atmospheric Administration's (NOAA's) Earth System Research Laboratory (ESRL) monitors CO<sub>2</sub> in the atmosphere as a contribution to the North American Carbon Program (NACP) (2). Mole fractions of CO<sub>2</sub> are determined with an accuracy of 0.1 parts per million (ppm) from surface air samples collected around the globe and from tall towers and small aircraft in North America. These measurements form a record of integrated net CO<sub>2</sub> exchange from multiple processes, geographic areas, and times.

In addition, carbon exchange is monitored locally ( $\approx 1$  km<sup>2</sup>) from a worldwide collection of surface flux measurements in different ecosystems and through periodic inventories of carbon in oceans, forests, and soils. The latter provide long-term constraints on the size of the different carbon pools. Monitoring of the carbon cycle through satellites mostly targets specific processes such as biomass burning, land-use change, or seasonal plant growth. Direct satellite observations of CO<sub>2</sub> are available already for the upper troposphere (3), whereas near-surface CO<sub>2</sub> from space will become available within several years to augment the current efforts.

To integrate this diversity of data into a consistent estimate of surface CO<sub>2</sub> exchange, the NOAA ESRL has built a new data assimilation system called CarbonTracker. It is used to retroactively analyze (reanalyze) the recent flux history of CO<sub>2</sub>, using a state-of-the-art atmospheric transport model coupled to an ensemble Kalman filter. Currently, CarbonTracker assimilates only atmospheric CO<sub>2</sub> mole fractions, but efforts to expand it to assimilate observations of other trace gases in the atmosphere (<sup>13</sup>CO<sub>2</sub>, <sup>14</sup>CO<sub>2</sub>, CH<sub>4</sub>) and other observation types (eddy-flux measurements, satellite radiances) are underway. Specifically, such observations could facilitate attribution of carbon fluxes to specific processes such as fossil fuel burning, biomass burning, or agricultural food and biofuel production.

One of the main innovations in CarbonTracker is the use of daily CO<sub>2</sub> values derived from continuous observations from a network of tall towers. These data were not available in similar previous studies (4–7) but are potentially highly informative on regional exchange patterns because they represent direct samples of the resulting strong gradients in space and time. The ability to use these data comes from the improved skill of our atmospheric transport model, the efficiency of the ensemble Kalman filter in solving large optimization problems, and the inclusion of subdaily variability in the surface flux models we try to optimize.

In this work, we introduce CarbonTracker and analyze the recent flux history it produces. We compare its regional estimates for North America with an independent “bottom-up” estimate that is part of the State of the Carbon Cycle Report (SOCCR) (8). This document, created as part of the U.S.

Author contributions: P.P.T. designed research; C.S., A.E.A., T.J.C., D.E.J.W., G.R.v.d.W., J.T.R., and P.O.W. contributed data; A.R.J., K.M., J.B.M., L.M.P.B., G.P., A.I.H., and M.C.K. performed research; and W.P. wrote the paper.

The authors declare no conflict of interest.

Freely available online through the PNAS open access option.

<sup>†</sup>To whom correspondence should be addressed. E-mail: [wouter.peters@noaa.gov](mailto:wouter.peters@noaa.gov).

This article contains supporting information online at [www.pnas.org/cgi/content/full/0708986104/DC1](http://www.pnas.org/cgi/content/full/0708986104/DC1).

© 2007 by The National Academy of Sciences of the USA

Climate Change Science Program and scheduled for release in the summer of 2007, is exclusively based on inventory data because atmosphere-based estimates were deemed too coarse and uncertain to report. We also assess CarbonTracker's skill in reproducing CO<sub>2</sub> observations from light aircraft not used in the assimilation, as an independent check on the realism of the estimated fluxes and vertical transport. This is specifically relevant for CarbonTracker's role in evaluating column average CO<sub>2</sub> observations that can be obtained from space-based sensors. CarbonTracker demonstrates the feasibility of monitoring the carbon cycle in substantial detail from high-quality atmospheric CO<sub>2</sub> observations.

### Assimilating CO<sub>2</sub>

The principles behind CarbonTracker are similar to other data assimilation systems. It starts by forecasting atmospheric CO<sub>2</sub> mole fractions around the globe from a combination of CO<sub>2</sub> surface exchange models and an atmospheric transport model driven by meteorological fields from the European Centre for Medium-Range Weather Forecasts (ECMWF). The resulting three-dimensional CO<sub>2</sub> distribution is then sampled at the time and location that observations are available, and the difference between observations and model forecast is minimized. This minimization is achieved by tuning a set of linear scaling factors that control the magnitude of the surface fluxes for larger, but subcontinental, geographical areas. Once the value of each of the scaling factors is determined in many consecutive assimilation cycles, the 6-yr history of surface CO<sub>2</sub> exchange at 1° × 1° can be readily constructed.

In addition to CO<sub>2</sub> mole fractions from all analyzed air samples from the NOAA ESRL Cooperative Air Sampling Network, we use daytime average mole fractions derived from continuous CO<sub>2</sub> time series at five towers [all calibrated against the world CO<sub>2</sub> standard (9)]: (i) the 396-m (above ground) level of the WLEF-TV tower near Park Falls, Wisconsin; (ii) the 107-m level of a cell phone tower near Argyle, Maine; (iii) the 457-m level of the KWKT-TV tower near Moody, Texas; (iv) the 40-m level of the tower in Fraserdale, Canada, operated by Environment Canada (EC); and (v) the 23-m level of the tower at Candle Lake, Canada, operated by EC. Other daytime average time series used are from the NOAA ESRL observatories at Barrow, Mauna Loa, Samoa, and the South Pole, and the continuous analyzer at Alert, Canada, operated by EC. The continuous data exhibit large variations on synoptic time scales resulting from the fact that changing wind directions bring different CO<sub>2</sub> signals to the sensors. These gradients, together with the modeled wind directions, inform the data assimilation system on regional flux differences. The total number of observations available (≈28,000) is small for data assimilation applications but the largest used in an atmosphere-based CO<sub>2</sub> estimate so far. Details on data treatment and a list of all of the sites is included in [supporting information \(SI\) Appendix](#).

Transport of atmospheric CO<sub>2</sub> is simulated by using the global two-way nested transport model TM5. TM5 is an offline model driven by 3- to 6-h meteorological parameters taken from the operational forecast of the ECMWF model (10). In this work, TM5 is run at a global 6° × 4° resolution with nested regions over North America (3° × 2°) and the United States (1° × 1°) (11). The choice of transport model is important in atmospheric CO<sub>2</sub> inverse modeling because the estimated fluxes were shown to be sensitive to vertical and horizontal transport (12). TM5 has been evaluated extensively and consistently performs well in ongoing intercomparisons. Possible biases in its vertical transport are assessed at the end of this article and in [SI Appendix](#).

We consider net CO<sub>2</sub> surface exchange from fossil fuel burning, fires, terrestrial biosphere exchange, and exchange with the oceans. Fossil fuel and fire emissions are fixed based on bottom-up estimates of their distribution and magnitude,

whereas biospheric and oceanic fluxes are adjusted to match the atmospheric CO<sub>2</sub> record. This choice reflects our faith in inventory-based emissions for fossil fuels and our lack of atmospheric observations to constrain tropical biomass burning CO<sub>2</sub> fluxes in this framework. Details on the applied surface fluxes are included in [SI Appendix](#). The four processes considered drive instantaneous CO<sub>2</sub> fluxes in the model according to

$$F(x, y, t) = \lambda_r \cdot F_{\text{bio}}(x, y, t) + \lambda_r \cdot F_{\text{occ}}(x, y, t) + F_{\text{ff}}(x, y, t) + F_{\text{fire}}(x, y, t), \quad [1]$$

where  $\lambda_r$  represents a set of linear scaling factors for each week and each region ( $r$ ) to be estimated in the assimilation. To create the spatial distribution of regions, the terrestrial biosphere is divided up according to ecosystem type and continent. Nineteen possible ecosystem types summarized in [SI Appendix](#) are therefore considered in each of 11 global land areas as in Gurney *et al.* (12), yielding a total of 25 ecoregions out of a possible 38 represented in boreal and temperate North America. The 13 ecosystem types not optimized for North America are either not represented (tropical forests in the boreal regions, for instance) or explicitly excluded based on physical considerations (e.g., deserts and polar ice caps were assumed to have zero carbon flux). Similarly, the ocean is divided into 11 large basins encompassing large-scale ocean circulation features. Thus, a total of  $r = 135$  scaling factors are optimized globally each week. The ensemble system used to solve for the scalar multiplication factors is similar to that in Peters *et al.* (13). Details of the ensemble Kalman filter design are discussed in [SI Appendix](#).

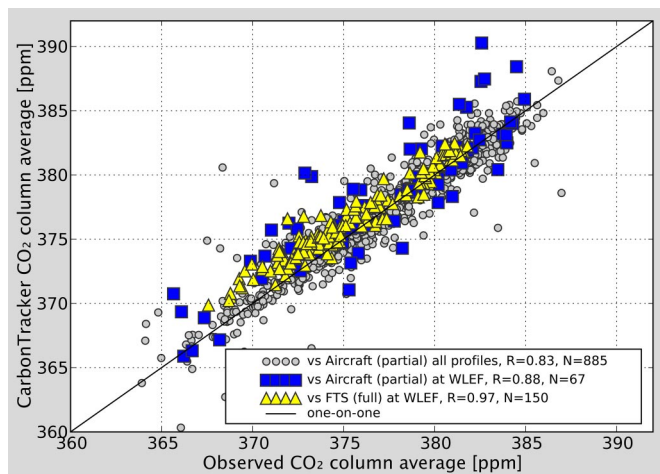
Relatively high temporal and spatial flux variations are represented in the fluxes  $F_{\text{bio}}(x, y, t)$  and  $F_{\text{occ}}(x, y, t)$  based on prescribed 3-h meteorological variables (e.g., sunlight, temperature, wind speed, surface pressure) and high-resolution input datasets (see [SI Appendix](#)) and not derived from atmospheric CO<sub>2</sub> observations. This recognizes the limited capacity of our network to sense CO<sub>2</sub> fluxes at the scales of individual counties or even states. Instead, we rely on mechanistic models to provide the high-frequency variations and optimize CO<sub>2</sub> fluxes over broader areas and time scales based on the available observations. The flux patterns at 1° × 1° shown in this work are thus the convolution of optimized parameters  $\lambda_r$  and fluxes  $F_{\text{bio}}(x, y, t)$  and  $F_{\text{occ}}(x, y, t)$ .

A cautionary note is that the high-resolution pattern prescribed to the system will influence the 1° × 1° results we present. Shortcomings in the high-resolution flux modules (such as the distribution of country total fossil fuel emissions by population density instead of using maps of power generation, energy consumption, and traffic density) are therefore also part of our final product. Although we have included specific sensitivity tests in [SI Appendix](#) to address some of these shortcomings, we caution anyone that the 1° × 1° results should not be interpreted without due consideration of the modeling framework chosen in this study.

Throughout the rest of this work, we will focus on results for North America (global flux estimates can be found in [SI Appendix](#)). We adopt the convention that negative fluxes denote a net loss of atmospheric CO<sub>2</sub> and a gain by a surface reservoir, and positive fluxes vice versa. Uncertainties are quoted as the upper and lower limits found in a set of 14 sensitivity experiments conducted to explore the possible outcomes given alternative choices for the main components of the system. This estimate thus addresses systematic modeling errors that were shown to exceed random estimation errors in previous studies (12, 14, 15). Although not quite a full Monte Carlo exploration, we feel that the set we have created forms a plausible range for the best estimate. Where appropriate, the best estimate will be followed by the minimum (subscript) and maximum (superscript) of the







**Fig. 5.** Comparison between column average CO<sub>2</sub> from observations and from our assimilated CO<sub>2</sub> distribution. Included are full column averages from FTS observations (yellow), partial column averages from aircraft samples at Parks Falls, Wisconsin (blue), and partial column averages from all NOAA ESRL aircraft observations (gray). Shown is the general good agreement between modeled CO<sub>2</sub> columns and independent (nonassimilated) observations. Linear correlation coefficients are given in the key. Units are mole fraction expressed as ppm.

the agreement on day-to-day variations and the amplitude of the seasonal cycle (see *SI Appendix*). This suggests that our model represents column average CO<sub>2</sub> well, at least at this location. For 22 other locations, we have integrated discrete samples from the NOAA ESRL Aircraft Program to give partial column average values for each flight (885 in total) and again cosampled our model to make a partial CO<sub>2</sub> column value. The excellent agreement ( $\Delta = 0.05 \pm 2.7$  ppm,  $R^2 = 0.83$ ) for this much larger domain (mostly North America and the eastern Pacific) and time span (2000–2006) is also shown in Fig. 5. For reference, partial columns for the WLEF-TV tower are included separately. Again, the comparison with independent data reveals no large biases in CarbonTracker fluxes and vertical transport as argued in more detail in *SI Appendix*.

The favorable comparison of our product to aircraft-derived column CO<sub>2</sub> and FTS-derived values shows a high level of consistency among all three. It also suggests that, at least over North America and the Northern Pacific, our three-dimensional CO<sub>2</sub> fields are a useful assimilation of sparse observational data that could be used to quantitatively assess satellite-derived CO<sub>2</sub> columns. In 2008, two dedicated missions called the Orbiting Carbon Observatory (OCO; National Aeronautics and Space Administration) and GoSat (Japanese Space Agency) will be launched for this purpose. Currently, the Atmospheric Infrared Sounder (AIRS) and the Scanning Imaging Absorption Spec-

trometer for Atmospheric Chartography (SCIAMACHY) already deliver experimental column average CO<sub>2</sub> products. Demands on the precision and accuracy of satellite-observed CO<sub>2</sub> are high (34, 35), and it is currently unclear whether the existing and planned satellite sensors can provide unbiased information on net carbon exchange. Our first analysis here suggests that flux and transport biases in CarbonTracker are small enough to evaluate such products quantitatively even at many locations where independent observations are not available.

### Discussion

The first release of CarbonTracker marks a significant step in our ability to monitor month-by-month surface sources and sinks of CO<sub>2</sub>. In addition to overcoming many technical hurdles, the system introduces a novel ensemble assimilation method and a large number of other innovations to model atmospheric CO<sub>2</sub> mole fractions accurately. CarbonTracker's ability to derive trustworthy surface fluxes depends strongly on the careful observations made by dedicated researchers all over the world. Expansion of the monitoring efforts to sparsely sampled regions of the world, and to collect more high-frequency time series across North America, should therefore have the highest priority. Moreover, independent monitoring of the isotopic fraction of <sup>14</sup>CO<sub>2</sub> in the atmosphere would allow independent verification of the fossil fuel fluxes by tuning for parameters in models describing those fluxes. Such estimates have great value for climate change mitigation policies. In addition, they would improve estimates of other fluxes because in the current version of CarbonTracker, relatively small errors in the fossil fuel emissions inventories may be aliased into relatively larger errors in other fluxes.

We stress that CarbonTracker currently is a framework on which to expand in future updates. Public release of new reanalyses incorporating both new data and innovations in the system itself are scheduled for October of each year starting in 2007. We welcome any contribution to CarbonTracker, and cooperation with those interested in the effort as CarbonTracker is intended to be a tool for the carbon cycle scientific community. In addition, we intend to improve and develop CarbonTracker further into a tool that is useful to policymakers from the national to the regional level. Therefore, all results, data, code, and other tools used in this study are freely available from NOAA ESRL's CarbonTracker web site, <http://carbontracker.noaa.gov>.

We thank Taro Takahashi, Greg Marland, T. J. Blasing, Tris West, Jerry Olson, and the EDGAR consortium for contributing data sets used to complete this study. Development of the GFED2 fire emissions was supported by National Aeronautics and Space Administration Grant NNG04GK496. The column CO<sub>2</sub> observations at Park Falls were obtained with support from National Aeronautics and Space Administration Grant NNG05GD07G. CarbonTracker is a National Oceanic and Atmospheric Administration contribution to the North American Carbon Program.

- Hinzman LD, Bettez ND, Bolton WR, Chapin FS, Dyurgerov MB, Fastie CL, Griffith B, Hollister RD, Hope A, Huntington HP, et al. (2005) *Clim Change* 72:251–298.
- Wofsy SC, Harriss RC (2002) *The North American Carbon Program (NACP) Report for the US Interagency Carbon Cycle Science Program* (US Interagency Carbon Cycle Science Program, Washington, DC).
- Engelen RJ, Stephens GL (2004) *J Appl Meteorol* 43:373–378.
- Fan S, Gloor M, Mahlman J, Pacala S, Sarmiento J, Takahashi T, Tans P (1998) *Science* 282:442–446.
- Rödenbeck C, Houweling S, Gloor M, Heimann M (2003) *Atmos Chem Phys* 3:1919–1964.
- Gurney KR, Law RM, Denning AS, Rayner PJ, Pak BC, Baker D, Bousquet P, Bruhwiler L, Chen YH, Ciais P, et al. (2004) *Global Biogeochem Cycles* 18:GB002111.
- Baker DF, Law RM, Gurney KR, Rayner P, Peylin P, Denning AS, Bousquet P, Bruhwiler L, Chen Y-H, Ciais P, et al. (2006) *Global Biogeochem Cycles* 20:GB002439.
- Pacala S, Birdsey RA, Conant RT, Davis K, Hales B, Jenkins JC, Johnston M, Marland G, Paustian K, Wofsy SC (2007) in *The First State of the Carbon Cycle Report (SOCCR): The North American Carbon Budget and Implications for the Global Carbon Cycle*, eds King AW, Dilling L, Zimmerman GP, Fairman DM, Houghton RA, Marland GA, Rose AZ, Wilbanks TJ (US Climate Change Science Program, Washington, DC), pp 69–91.
- World Meteorological Organization (2007) *Global Atmosphere Watch Report*, ed Miller JB (World Meteorological Organization, Geneva), no 168.
- Krol MC, Houweling S, Bregman B, van den Broek M, Segers A, van Velthoven P, Peters W, Dentener FJ, Bergamaschi P (2005) *Atmos Chem Phys* 5:417–432.
- Peters W, Krol MC, Dlugokencky E, Dentener FJ, Bergamaschi P, Dutton G, van Velthoven P, Miller JB, Bruhwiler L, Tans PP (2004) *J Geophys Res Atmos* 109:JD005020.
- Gurney KR, Law RM, Denning AS, Rayner PJ, Baker D, Bousquet P, Bruhwiler L, Chen YH, Ciais P, Fan S, et al. (2002) *Nature* 415:626–630.

13. Peters W, Miller JB, Whitaker J, Denning AS, Hirsch A, Krol MC, Zupanski D, Bruhwiler L, Tans PP (2005) *J Geophys Res Atmos* 110:JD006157.
14. Xiao X (2007) *J Geophys Res Atmos* 112:JD07303.
15. Peylin P, Baker D, Sarmiento J, Ciais P, Bousquet P (2002) *J Geophys Res Atmos* 107:JD000857.
16. Olson JS, Watts JA, Allison LJ (1985) *Technical Report NDP-017* (Oak Ridge National Laboratory, Oak Ridge, TN).
17. Houghton RA, Hackler JL (1999) *Global Change Biol* 5:481–492.
18. Caspersen JP, Pacala SW, Jenkins JC, Hurtt GC, Moorcroft PR, Birdsey RA (2000) *Science* 290:1148–1151.
19. Pacala SW, Hurtt GC, Baker D, Peylin P, Houghton RA, Birdsey RA, Heath L, Sundquist ET, Stallard RF, Ciais P, et al. (2001) *Science* 292:2316–2320.
20. Hurtt GC (2002) *Proc Natl Acad Sci USA* 99:1389–1394.
21. Ciais P, Bousquet P, Freibauer A, Naegler T (2007) *Global Biogeochem Cycles* 21:GB002741.
22. US Environmental Protection Agency (2005) *Report 430-R-05-003* (US Environmental Protection Agency, Washington, DC).
23. Masera OR (1997) *Clim Change* 35:265–295.
24. Skog KE (2004) *Environ Manage* 33:S65–S73.
25. Zeng N, Mariotti A, Wetzel P (2005) *Global Biogeochem Cycles* 19:GB002273.
26. Chen JM, Chen B, Higuchi K, Liu J, Chan D, Worthy D, Tans P, Black A (2006) *Geophys Res Lett* 33:GL025919.
27. Ciais P, Viovy N, Chevallier F, De Noblet N, Friend AD, Friedlingstein P, Reichstein M, Manca G, Papale D, Valentini R, et al. (2005) *Nature* 437:529–533.
28. van der Werf GR, Randerson JT, Giglio L, Collatz GJ, Kasibhatla PS, Arellano AF, Jr (2006) *Atmos Chem Phys* 6:3423–3441.
29. Zeng N, Qian HF, Rödenbeck C, Heimann M (2005) *Geophys Res Lett* 32:GL024607.
30. Stephens BB, Gurney KR, Tans PP, Sweeney C, Peters W, Bruhwiler L, Ciais P, Ramonet M, Bousquet P, Nakazawa T, et al. (2007) *Science* 316:1732–1735.
31. Yang Z, Washenfelder RA, Keppel-Aleks G, Krakauer NY, Randerson JT, Tans PP, Sweeney C, Wennberg PO (2007) *Geophys Res Lett* 34:GL029742.
32. Bosch H, Toon GC, Sen B, Washenfelder RA, Wennberg PO, Buchwitz M, de Beek R, Burrows JP, Crisp D, Christi M, et al. (2006) *J Geophys Res Atmos* 111:D23302.
33. Washenfelder RA, Toon GC, Blavier JF, Yang Z, Allen NT, Wennberg PO, Vay SA, Matross DM, Daube BC (2006) *J Geophys Res Atmos* 111:D22305.
34. Rayner PJ, O'Brien DM (2001) *Geophys Res Lett* 28:175–178.
35. Houweling S, Breon FM, Aben I, Rödenbeck C, Gloor M, Heimann M, Ciais P (2004) *Atmos Chem Phys* 4:523–538.