Emissions of greenhouse gases from a North American megacity

D. Wunch,[1] P. O. Wennberg,[1] G. C. Toon,[1,2] G. Keppel-Aleks,[1] and Y. G. Yavin[1,3]

Received 29 June 2009; revised 8 July 2009; accepted 9 July 2009; published 6 August 2009.

[1] Atmospheric column abundances of carbon dioxide (CO_2), carbon monoxide (CO), methane (CH_4) and nitrous oxide (N_2O) have been measured above the South Coast air basin (SCB), a densely populated urban region of Southern California, USA, which includes Los Angeles and the surrounding suburbs. Large diurnal variations in CO and CH_4 are observed which correlate well with those in CO_2. Weaker correlations are seen between N_2O and CO_2, with large uncertainties. We compute yearly SCB emissions of CO and CH_4 to be 1.4 ± 0.3 Tg CO and 0.6 ± 0.1 Tg CH_4. We compare our calculated emissions to the California Air Resources Board (CARB) and the Emission Database for Global Atmospheric Research (EDGAR) estimates. Our measurements confirm that urban emissions are a significant source of CH_4 and in fact may be substantially higher than currently estimated. If our emissions are typical of other urban centers, these findings suggest that urban emissions could contribute 7–15% to the global anthropogenic budget of methane. **Citation:** Wunch, D., P. O. Wennberg, G. C. Toon, G. Keppel-Aleks, and Y. G. Yavin (2009), Emissions of greenhouse gases from a North American megacity, *Geophys. Res. Lett.*, *36*, L15810, doi:10.1029/2009GL039825.

1. Introduction

[2] Methane (CH_4), nitrous oxide (N_2O) and carbon dioxide (CO_2) are long-lived greenhouse gases that are primary contributors to climate change. Carbon monoxide (CO), although a weak direct greenhouse gas, is considered an ozone precursor gas and a pollutant [*Solomon et al.*, 2007]. Both CO_2 and CO are known to be emitted in significant amounts in urban areas. The main sources of the CO_2 emissions are fossil fuel use, with a smaller but significant source due to land use changes [*Denman et al.*, 2007]. CO is a product of incomplete combustion, and its main global sources are biomass burning, fossil fuel combustion (including passenger vehicles [*California Air Resources Board* (*CARB*), 2008]), agricultural waste burning, biofuel combustion and industrial processes (§1.7.4.2 [*Montzka et al.*, 2003]).

[3] While the urban sources of CO_2 and CO have been extensively studied, there has been a paucity of observational studies of CH_4 and N_2O emissions in urban environments (§2.3.2 [*Forster et al.*, 2007]).

[4] The total global CH_4 emissions are known relatively well (582 ± 50 Tg/yr), but the strengths of individual sources are not (§2.3.2 [*Forster et al.*, 2007]). Current estimates suggest that 70% of the global source of methane is biogenic, coming from methanogens present in wetlands, rice paddies, ruminants, landfills, oceans and forests. Non-biogenic sources include fossil fuel mining and burning, biomass burning, waste treatment and geological sources (§7.4.1 [*Denman et al.*, 2007]).

[5] The total global N_2O emissions are between 26.7 and 87 Tg/yr; 38% of the source is thought to be anthropogenic [*Denman et al.*, 2007, Table 7.7]. Of the anthropogenic sources, the largest is agriculture (42%), followed by oceanic/estuary/river sources (25%), biomass and biofuel burning (10%), fossil fuel burning (10%) and atmospheric deposition (9%). N_2O is also a known product of vehicle exhaust [*Becker et al.*, 2000].

[6] The IPCC reports large ranges in the anthropogenic emissions of CH_4 and N_2O that could be partially attributable to urban regions. CH_4 produced by gas and oil production, industry, landfills and waste treatment accounts for 15% to 40% of global anthropogenic CH_4 emissions [*Denman et al.*, 2007]. Urban sources of N_2O could include fossil fuel burning (transportation), fertilizer use and industrial production and are estimated to account for 1% to 10% of the global N_2O emissions [*Denman et al.*, 2007].

[7] Data described in this paper, recorded at the Jet Propulsion Laboratory (JPL), in California, USA, show strong diurnal variations in CH_4 and weaker ones in N_2O which are associated with the local urban emissions. The enhancements in methane are highly correlated with those in carbon monoxide and carbon dioxide. If the correlations are typical of urban areas worldwide, these data suggest that urban areas contribute more significantly to the global methane budget than currently thought.

2. Methods

[8] Atmospheric CO_2, CO, CH_4 and N_2O are measured with a ground-based Fourier transform spectrometer (FTS) that records the near infrared (NIR) spectrum of the direct solar beam that has passed through the atmosphere. A solar tracker allows measurements of spectra throughout the day, and thus information on the diurnal behavior of the gases is obtained. From these spectra, we retrieve the vertically-integrated total columns of CO_2, CO, CH_4 and N_2O with the non-linear least squares spectral fitting algorithm GFIT, which was developed at JPL. The columns allow surface fluxes to be estimated with little influence from the diurnal changes in the boundary layer thickness [*Gloor et al.*, 2000]. To remove the effects of surface pressure variation, column-averaged dry-air mole fractions (DMF), denoted for gas G by xG, are computed by dividing the columns by the column of dry air, which we derive from the O_2 column measured in the same spectra. Details of the retrieval

[1]Division of Geological and Planetary Sciences, California Institute of Technology, Pasadena, California, USA.
[2]Jet Propulsion Laboratory, California Institute of Technology, Pasadena, California, USA.
[3]Now at 5 Granot St., Jerusalem, Israel.

0094-8276/09/2009GL039825$05.00

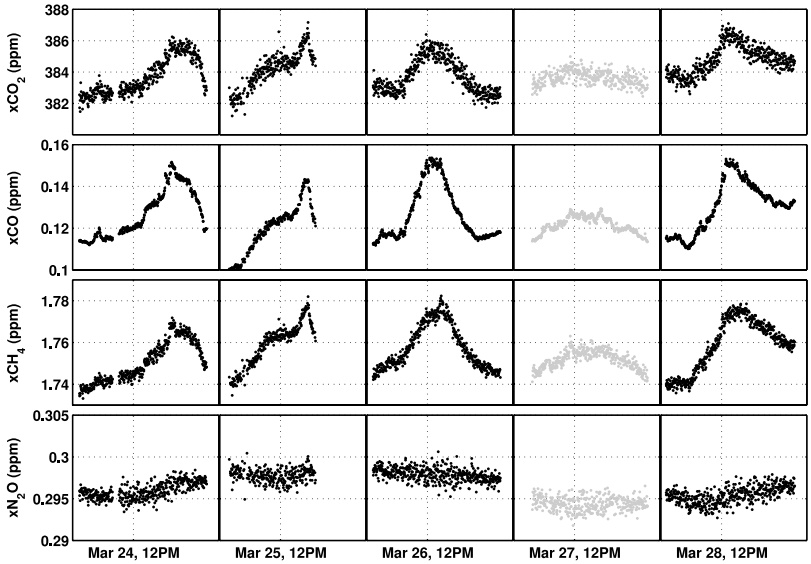

Figure 1. The four plots show the xCO_2, xCO, xCH_4 and xN_2O over the course of five days. For March 24–26 and March 28, the xCO_2, xCO and xCH_4 show strong diurnal variation associated with SCB emissions. The March 27 data show much smaller variations, as the air originated from the Mojave desert. Note that there is no data recorded at night, and the solid vertical lines represent the time between 7 PM and 7 AM.

method are given in Retrievals and Spectroscopy of Text S1 and by *Washenfelder et al.* [2006].[1]

[9] The instrument was located at JPL (34.2 N, 118.2 W, 390 masl), near Los Angeles, California, USA from August, 2007 to June, 2008. JPL is situated at the northern limit of the South Coast air basin (SCB). Due to the large population of the SCB (15 million [*California State Data Center*, 2003]) and the basin being bounded on three sides by mountains and by the Pacific Ocean on the fourth, the SCB contains some of the most polluted air in the USA. Under the normal prevailing meteorological conditions with winds from the west or southwest, air reaching JPL is polluted. The area to the north and east of the air basin is the sparsely populated Mojave desert. Occasionally, with winds from the north or east, JPL receives clean air.

[10] The FTS time series of xCO_2, xCO, xCH_4 and xN_2O show slowly-varying changes (see Figure 1 of S1). In addition to these slow changes, significant diurnal variability is observed. Data from March 24–26 and 28, 2008 show diurnal changes due to activity in the SCB, whereas the March 27 data show the relatively clean air that originated from the north (Figure 1), according to the HYSPLIT back-trajectory model [*Rolph*, 2003; *Draxler and Rolph*, 2003]. To confine the analysis to emissions within the basin, all days for which the xCO_2 changes by less than 2 ppm are excluded. We also exclude data from days affected by wildfires, as those days contain large CO contributions that are not associated with local urban emissions. Of the 268 days of measurements, 131 days were included in this analysis (see Data Filtering of Text S1 for details).

[11] The diurnal variations of xCO_2, xCO, xCH_4 and xN_2O are highly correlated (Figure 2 and Table 1). The correlations (and errors) were determined by linear regression of the data shown in Figure 2 using the *York et al.* [2004] method that takes into account errors in both the abscissa and ordinate values. The data shown are daily anomalies, computed by subtracting the morning DMF at a particular solar zenith angle (SZA) from its afternoon counterpart. This method eliminates the possibility that SZA-dependent errors (e.g. due to spectroscopic inadequacies) cause spurious correlations. We assume in this analysis that the observed diurnal changes are confined to the boundary layer, and so the anomalies have been divided by the averaging kernel value at the surface to account for the sensitivity of the column measurement on variations at the surface.

3. Results

[12] The correlations between the trace gas columns arise from diurnal changes in the polluted urban basin air as

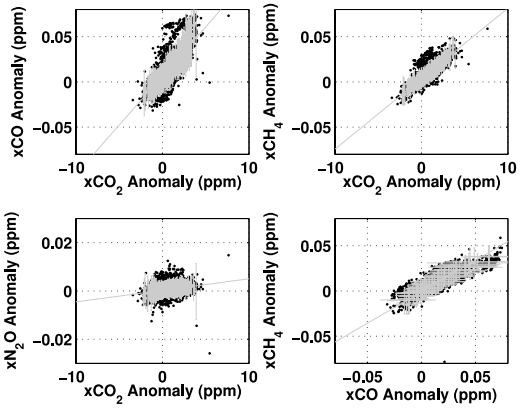

Figure 2. Correlations of the anomalies of xCO_2, xCO, xCH_4 and xN_2O. The grey points are the means for each 0.1 ppm xCO_2 anomaly interval, and the error bars represent two standard deviations on the mean.

[1]Auxiliary materials are available in the HTML. doi:10.1029/2009GL039825.

Table 1. Slopes of the Anomaly Correlations of xCO, xCH_4 and xN_2O to xCO_2 from Figure 2 in mol/mol, and the Mass Ratio of the Molecules to CO_2

	α_G (per mil)	Mass Ratio (M_G/M_{CO_2})
xCO	11 ± 2	28/44
xCH_4	7.8 ± 0.8	16/44
xN_2O	0.5 ± 0.3	44/44

observed at JPL. These diurnal signals are not prominent in air that has come from the desert to the north or east (e.g., Figure 1), nor in data from similar FTS instruments in clean-air locations, such as Park Falls, Wisconsin, USA and Darwin, Australia. The correlations are due to the buildup of pollutants in the SCB through the morning, while the planetary boundary layer (PBL) height is below the tops of the surrounding mountains [*Ulrickson and Mass*, 1990]. On most days, a decrease is observed in the xCO_2, xCO and xCH_4 in the late afternoon, again due to the dynamics of the SCB (Figure 1). Anomalies in xCO_2 of as much as 8 ppm are observed, representing a 2% change in the total column abundance. This is in reasonable agreement with the expected daily CO_2 emissions of 1% of the total column abundance in the SCB (see Diurnal Variation of Text S1 for calculation details).

[13] If the total emissions of any of CO_2, CO or CH_4 are known for the SCB, the emissions of the other gases can be estimated by multiplying by the appropriate correlation slopes and the molecular mass ratios. Using the correlation slopes is a better choice than using the anomalies themselves, because correlations are independent of transport and other atmospheric effects within the SCB that are common to both gases. For example, if the emission of CO_2 in the SCB is known, the emission of gas G (any of CO, CH_4 or N_2O) is:

$$E_G^{SCB} = \left(\alpha_G \frac{M_G}{M_{CO_2}}\right) E_{CO_2}^{SCB}, \quad (1)$$

where E_G^{SCB} is the emission from the SCB from gas G in Tg G, α_G is the correlation slope of gas G to xCO_2 in mol/mol, M_G is the molecular mass of gas G in g/mol and $E_{CO_2}^{SCB}$ is the SCB CO_2 emissions in Tg CO_2.

[14] This analysis assumes that the *diurnal* dependence of the emissions for all of CO_2, CO, CH_4 and N_2O are similar. Thus, the correlations shown in Figure 2 should not be interpreted to imply that the sources are common between xCO, xCH_4, xN_2O and xCO_2. Methane, for example, has sources that are relatively constant diurnally (landfills and natural gas leakage), whereas the emissions of CO are primarily from traffic, which have stronger daytime sources. If we were to compute CH_4 emissions from CO emissions, using equation (1), our analysis may underestimate the CH_4 emissions.

4. Discussion

[15] The California Air Resources Board (CARB), publishes state-wide inventories of greenhouse gas emissions [*CARB*, 2006]. CARB estimates that the state-wide emissions of CO_2 in 2006 were 444 ± 67 Tg CO_2, levels which have been stable to within ~5% since 2000. (We assume a 15% error on the estimates and do not include emissions from imported electricity generation.)

[16] CARB also provides CO emissions estimates for the SCB (1.1 ± 0.2 Tg CO) and for California (3.8 ± 0.6 Tg CO) for 2008 [*CARB*, 2008]. The CARB CO emissions exclude those produced from wildfires. The CARB state-wide estimates of CO_2, CH_4, N_2O and CO emissions are listed in Table 2.

[17] In addition to CARB, the Emission Database for Global Atmospheric Research (EDGAR) [*Olivier et al.*, 1994, 2005; *EDGAR Project Team*, 2009] is often used in emissions studies. EDGAR is a gridded inventory for global CO_2, CO, CH_4 and N_2O emissions, as well as other greenhouse gases and precursor gases (see EDGAR Grid of Text S1). EDGAR currently provides CO_2, CH_4 and N_2O estimates for 2005, and CO for 2000. The total CO_2 emissions from EDGAR for the state of California are 490 ± 74 Tg CO_2, in good agreement with the CARB value.

[18] To compute the CARB SCB CO_2 emissions, the CO_2 emissions are assumed to scale with population, and so the state-wide CARB CO_2 emissions are multiplied by the fraction of Californians living in the SCB (43% [*California State Data Center*, 2003]). This is consistent with the EDGAR model, where CO_2 attributed to the SCB is 41% of the state total.

[19] Despite the good agreement between CARB and EDGAR CO_2 emissions, the EDGAR state-wide emissions of CO and CH_4 are significantly larger than the CARB inventories. The CARB CO emission for 2000 is 6.4 Tg/yr, 2/3 the EDGAR California estimate (9.8 Tg/yr). The CARB 2006 CH_4 emission is 1.3 ± 0.2 Tg/yr, compared with 2.3 ± 0.3 Tg/yr from EDGAR 2005. The CARB 2006 N_2O emission, 0.0046 ± 0.007 Tg/yr, is in good agreement with the EDGAR 2005 N_2O, 0.049 ± 0.007 Tg/yr. As discussed below, we believe the differences result from how the EDGAR US emissions are desegregated to local regions.

[20] Because the CO_2 emissions are less variable from year to year than the CO emissions, the SCB CO_2 emissions are used to compute FTS-derived emissions of CO, CH_4 and N_2O using equation (1). Since the CARB and EDGAR SCB CO_2 results agree within error, the average is used: $E_{CO_2}^{SCB}$ = 198 ± 30 Tg/yr CO_2. The FTS-derived SCB emissions computed from equation (1) and the slopes in Table 1 are listed in Table 2. The errors in the slopes were propagated to compute the errors on the emissions listed in Table 2.

4.1. Carbon Monoxide

[21] Emissions of CO inferred here, 1.4 ± 0.3 Tg/yr, are in good agreement with the CARB inventory (1.1 ± 0.2 Tg/yr), but in poor agreement with EDGAR, whose emissions for 2000 are 4.4 Tg/yr. Accounting for a 5.5% drop in SCB CO emissions per year for 7.5 years, the 2007–2008 EDGAR emissions estimates become 2.6 Tg/yr, over-estimating the emissions by about a factor of two. This over-estimate is likely due to EDGAR's method of producing gridded CO data: distributing US aggregated on-road emissions by road density. California's CO emissions regulations are the strictest in the country, and so have much lower emissions than the EDGAR method would predict.

Table 2. California Emissions and South Coast Air Basin (SCB) Emissions from CARB and EDGAR in Tg/yr, where available, and FTS-derived[a] SCB Emissions in Tg/yr

	California Emissions				SCB Emissions				
	CARB		EDGAR		CARB		EDGAR		FTS
	2006	2008	2000	2005	2006	2008	2000	2005	2007–2008
CO_2	444	—	—	490	191	—	—	206	—
CO	—	3.76	9.80	—	—	1.08	4.35	—	1.4 ± 0.3
CH_4	1.30	—	—	2.26	—	—	—	0.69	0.6 ± 0.1
N_2O	0.046	—	—	0.049	—	—	–	0.015	0.10 ± 0.06

[a]Using the data from Table 1 and equation (1), the FTS-derived CO, CH_4 and N_2O emissions in the SCB are estimated from the average of the CARB and EDGAR values for the SCB CO_2 (198 ± 30 Tg).

4.2. Methane

[22] The CARB inventory suggests that 55% of the state's CH_4 emissions come from the Agriculture and Forestry sector, while the SCB contains less than 5% of California's farmland [*United States Department of Agriculture*, 2002]. Removing the agriculture and forestry sources of CH_4 from the CARB California CH_4 emissions gives "other" emissions, primarily from landfills, wastewater treatment and pipelines, of 0.6 Tg/yr. Assuming the emissions scale with population, we infer that CARB's urban CH_4 inventory for the SCB is ~0.26 Tg/yr — less than half the flux determined here. Alternatively, if the CARB SCB CO emissions are used to compute the CH_4 flux, we can calculate a lower bound, 0.4 ± 0.1 Tg/yr, which remains significantly larger than the inventory.

[23] What sources might be responsible for the 0.14–0.34 Tg/yr unaccounted SCB CH_4 emissions? Since much of the SCB is powered by natural gas, a possible source of the atmospheric CH_4 in the SCB may be from unaccounted gas leaks. According to California's Energy Consumption and Data Management System [*Energy Consumption Data Management System*, 2006], the SCB consumed 10.5 Tg CH_4 in 2006. This would represent a 1%–3% loss in the natural gas delivery system in addition to CARB's pipeline loss estimate of 0.091 Tg/yr state-wide. This is plausible when compared with the ~1% that previous studies by *Lelieveld et al.* [2005] suggest.

[24] The FTS-derived SCB CH_4 emissions are in good but fortuitous agreement with the EDGAR SCB emissions. The main discrepancy between the EDGAR and CARB CH_4 emissions is the amount produced by waste. CARB reports CH_4 produced by California landfills and wastewater treatment to be 0.3 Tg/yr, and EDGAR reports Californian "waste" emissions of 1.3 Tg/yr. However, EDGAR gridded waste emissions are derived from aggregate US emissions, distributed by population density. This method does not account for either regional differences in landfill gas production, or recapture, for which California is at the forefront. This will spuriously inflate the EDGAR California emissions from landfills. It would be possible to test whether the excess CH_4 in the SCB is derived from landfills/biogenics or natural gas leaks by measuring the $^{14}C/^{12}C$ amounts in CH_4, as natural gas-derived CH_4 will have no ^{14}C.

[25] If the same correlation coefficients in Table 1 are found in other large urban areas worldwide, then the global anthropogenic emissions of CO_2 should give a good estimate of the global urban CH_4 emissions. The IPCC estimated 2007 global CO_2 emissions to be 31,450 Tg CO_2/year [*Marland et al.*, 2008; *Denman et al.*, 2007]. If we assume, as an upper limit, that all anthropogenic emissions of CO_2 originate from urban areas, then this number leads to a global urban CH_4 emission of 89 ± 9 Tg CH_4/yr, or 21–34% of the estimates of the total global anthropogenic emissions (264–428 Tg CH_4/yr) [see *Denman et al.*, 2007, Table 7.6]. The unaccounted CH_4 in the SCB, 0.2 ± 0.1 Tg/yr, scales to 40 ± 20 Tg/yr globally by urban population [*United Nations Population Division*, 2009], and 32 ± 16 Tg/yr globally by CO_2 emissions, which is 7%–15% of the anthropogenic total. Since the sources of CH_4 in the SCB are unknown, these global estimates carry a high degree of uncertainty.

4.3. Nitrous Oxide

[26] The FTS-derived SCB N_2O emissions are highly uncertain but larger than the CARB and EDGAR state-wide N_2O estimates. Furthermore, the CARB inventory suggests that the Agriculture and Forestry sector accounts for about 60% of the state-wide N_2O, which would make the FTS-derived SCB N_2O emissions between 2 and 8 times larger than the CARB state-wide urban N_2O emissions. Given the large uncertainty, it is difficult to draw any strong conclusions about N_2O emissions.

5. Summary and Conclusions

[27] Correlations derived from FTS column measurements of CO_2, CH_4 and CO are used to compute emissions of CH_4 and CO within the SCB. The computed emissions of CH_4 reveal an underestimate of the urban CH_4 emissions by CARB. The computed CO emissions are in good agreement with CARB CO estimates. Compared with the computed emissions, EDGAR is in fortuitously good agreement with the emissions of CH_4, and overestimates emissions of CO in the SCB. The same analysis applied to global CO_2 emissions reported by the IPCC show that urban CH_4 emissions may account for 21–34% of the total global anthropogenic CH_4 emissions, with the unaccounted emissions adding 7%–15% to the global budget.

[28] Our analysis does not identify the sources of SCB emissions of any of the gases analyzed. To do so, in situ samples, including isotopic analysis, in different areas of the basin would be required. Similar analysis in other air basins would help determine the robustness of the correlation coefficients and may also help isolate emissions sources.

[29] **Acknowledgments.** The authors thank Alan Kwan, Kathleen Spencer, Amy Townsend-Small, Charles E. Miller and Ying K. Hsu for helpful discussions, Jean-François L. Blavier for invaluable technical assistance and two anonymous reviewers for insightful comments. The authors gratefully acknowledge the NOAA Air Resources Laboratory

(ARL) for the provision of the READY website (http://www.arl.noaa.gov/ready.html) used in this publication [*Draxler and Rolph*, 2003; *Rolph*, 2003]. Part of this research was conducted at the Jet Propulsion Laboratory, California Institute of Technology, under contract with NASA. Support for this work is from the NASA Carbon Cycle Program grant NNX08AI86G and from the NASA OCO Project grant NAS7-03001.

References

Becker, K., J. Lörzer, R. Kurtenbach, P. Wiesen, T. Jensen, and T. Wallington (2000), Contribution of vehicle exhaust to the global N_2O budget, *Chemosphere Global Change Sci.*, *2*(3–4), 387–395.

California Air Resources Board (CARB) (2006), Greenhouse Gas Inventory Data—2000 to 2006, technical report, Calif. Air. Resour. Board, Sacramento. (Available at http://www.arb.ca.gov/app/ghg/2000_2006/ghg_sector.php)

California Air Resources Board (CARB) (2008), California Emission Inventory Data Almanac, technical report. (Available at http://www.arb.ca.gov/app/emsinv/emssumcat.php)

California State Data Center (2003), Census 2000 Summary File 4, technical report, U.S. Census Bur., Washington, D. C. (Available at http://quickfacts.census.gov/qfd/states/06000.html)

Denman, K., et al. (2007), Couplings between changes in the climate system and biogeochemistry, in *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by S. Solomon et al., pp. 499–587, Cambridge Univ. Press, Cambridge, U. K.

Draxler, R., and G. Rolph (2003), HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) Model access via NOAA ARL READY Website, Air. Resour. Lab., NOAA, Silver Spring, Md. (Available at http://www.arl.noaa.gov/ready/hysplit4.html)

EDGAR Project Team (2009), Emission Database for Global Atmospheric Research (EDGAR), release version 4.0., http://edgar.jrc.ec.europa.eu/index.php, Eur. Comm. Joint Res. Cent., Brussels.

Energy Consumption Data Management System (2006), Energy Consumption Data Management Service—California natural gas consumption, http://ecdms.energy.ca.gov/, Calif. Energy Comm., Sacramento.

Forster, P., et al. (2007), Changes in atmospheric constituents and in radiative forcing, in *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by S. Solomon et al., pp. 129–234, Cambridge Univ. Press, Cambridge, U. K.

Gloor, M., S.-M. Fan, S. Pacala, and J. Sarmiento (2000), Optimal sampling of the atmosphere for purpose of inverse modeling: A model study, *Global Biogeochem. Cycles*, *14*(1), 407–428.

Lelieveld, J., S. Lechtenböhmer, S. Assonov, C. Brenninkmeijer, C. Dienst, M. Fischedick, and T. Hanke (2005), Low methane leakage from gas pipelines, *Nature*, *434*(7035), 841–842.

Marland, G., T. Bode, and R. Andres (2008), Global, regional, and national CO_2 emissions, in *Trends: A Compendium of Data on Global Change*, technical report, Carbon Dioxide Inf. Anal. Cent., Oak Ridge Natl. Lab., Oak Ridge, Tenn.

Montzka, S., et al. (2003), Controlled substances and other source gases, in *Scientific Assessment of Ozone Depletion: 2002*, Global Ozone Res. Monit. Proj. Rep. 47, pp. 1.1–1.83, World Meteorol. Organ., Geneva, Switzerland.

Olivier, J., A. Bouwman, C. Maas, and J. Berdowski (1994), Emission Database for Global Atmospheric Research (EDGAR), *Environ. Monit. Assess.*, *31*(1), 93–106.

Olivier, J., J. Van Aardenne, F. Dentener, V. Pagliari, L. Ganzeveld, and J. Peters (2005), Recent trends in global greenhouse gas emissions: Regional trends 1970–2000 and spatial distribution of key sources in 2000, *Environ. Sci.*, *2*(2), 81–99.

Rolph, G. (2003), Real-time Environmental Applications and Display sYstem (READY), Air. Resour. Lab., NOAA, Silver Spring, Md. (Available at http://www.arl.noaa.gov/ready/hysplit4.html)

Solomon, S., D. Qin, M. Manning, Z. Chen, M. Marquis, K. Averyt, M. Tignor, and H. Miller (Eds.) (2007), *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, Cambridge Univ. Press, Cambridge, U. K.

Ulrickson, B., and C. Mass (1990), Numerical investigation of mesoscale circulations over the Los Angeles Basin. Part I. A verification study, *Mon. Weather Rev.*, *118*(10), 2138–2161.

United Nations Population Division (2009), World Urbanization Prospects: The 2007 Revision Population Database, http://esa.un.org/unup/, U. N., New York, N. Y.

United States Department of Agriculture (2002), The Census of Agriculture: 2002 Census Publications, technical report, U.S. Dep. Agric., Washington, D. C. (Available at http://www.agcensus.usda.gov/Publications/2002/Census_by_State/California/index.asp)

Washenfelder, R. A., G. C. Toon, J.-F. Blavier, Z. Yang, N. T. Allen, P. O. Wennberg, S. A. Vay, D. M. Matross, and B. C. Daube (2006), Carbon dioxide column abundances at the Wisconsin Tall Tower site, *J. Geophys. Res.*, *111*, D22305, doi:10.1029/2006JD007154.

York, D., N. Evensen, M. Martínez, and J. Delgado (2004), Unified equations for the slope, intercept, and standard errors of the best straight line, *Am. J. Phys.*, *72*(3), 367–375.

G. Keppel-Aleks, P. O. Wennberg, and D. Wunch, Division of Geological and Planetary Sciences, California Institute of Technology, M/C 150-21, 1200 East California Boulevard, Pasadena, CA 91125, USA. (dwunch@gps.caltech.edu)

G. C. Toon, Jet Propulsion Laboratory, California Institute of Technology, 4800 Oak Grove Drive Pasadena, CA 91109, USA.

Y. G. Yavin, 5 Granot Street, Jerusalem 91046, Israel.