



Is the airborne fraction of anthropogenic CO₂ emissions increasing?

Wolfgang Knorr¹

Received 18 August 2009; revised 21 September 2009; accepted 23 September 2009; published 7 November 2009.

[1] Several recent studies have highlighted the possibility that the oceans and terrestrial ecosystems have started losing part of their ability to sequester a large proportion of the anthropogenic CO₂ emissions. This is an important claim, because so far only about 40% of those emissions have stayed in the atmosphere, which has prevented additional climate change. This study re-examines the available atmospheric CO₂ and emissions data including their uncertainties. It is shown that with those uncertainties, the trend in the airborne fraction since 1850 has been $0.7 \pm 1.4\%$ per decade, i.e. close to and not significantly different from zero. The analysis further shows that the statistical model of a constant airborne fraction agrees best with the available data if emissions from land use change are scaled down to 82% or less of their original estimates. Despite the predictions of coupled climate-carbon cycle models, no trend in the airborne fraction can be found. **Citation:** Knorr, W. (2009), Is the airborne fraction of anthropogenic CO₂ emissions increasing?, *Geophys. Res. Lett.*, 36, L21710, doi:10.1029/2009GL040613.

1. Introduction

[2] Of the current 10 billion tons of carbon (GtC) emitted annually as CO₂ into the atmosphere by human activities [Boden *et al.*, 2009; Houghton, 2008], only around 40% [Jones and Cox, 2005] remain in the atmosphere, while the rest is absorbed by the oceans and the land biota to about equal proportions [Bopp *et al.*, 2002]. This airborne fraction of anthropogenic CO₂ (AF) is known to have stayed remarkably constant over the past five decades [Jones and Cox, 2005], but if it were to increase in a way predicted by models, this could add another 500 ppm of CO₂ to the atmosphere by 2100 [Friedlingstein *et al.*, 2006], significantly more than the current total. While recent studies have highlighted a decreasing ability of the Earth system to absorb the excess CO₂ [Le Quéré *et al.*, 2007; Schuster and Watson, 2007; Canadell *et al.*, 2007], the question if and why the airborne fraction has stayed constant at the decadal time scale has received less attention.

[3] Previous studies have found an AF of $40 \pm 14\%$ [Jones and Cox, 2005], and a slight upward trend of the AF of $2.5 \pm 2.1\%$ per decade [Canadell *et al.*, 2007]. While the trend was found to be significant at a 89% level, the analysis accounted only for the error inherent in the linear model, not for uncertainties in either the emissions or the atmospheric growth rate. A subsequent analysis by Raupach *et al.* [2008] has tested the robustness of this result against different assumptions about land use emissions and found that for

one of them, the estimated trend reduces to zero. The analysis, however, does not propagate the error in the land use flux to the uncertainty in the trend estimate, nor does it include data from before the late 1950s, when direct CO₂ measurements began. According to Etheridge *et al.* [1996], between 1850 and 1960 atmospheric CO₂ increased by 66 GtC (from 285 to 316 ppm, with 2.13 GtC per ppm [Trenberth, 1981]), which is 41% of the total emissions of 162 GtC during the same period [Boden *et al.*, 2009; Houghton, 2008].

[4] The purpose of the present study is to extend the trend analysis of the AF back to 1850, to re-do the analysis with data uncertainties and to re-examine the use of predictors of interannual variability. The results of several approaches are compared to re-consider the significance of any observed trend in the AF.

2. Data

[5] The present analysis combines the average concentration at Mauna Loa [Keeling *et al.*, 2009] and South Pole [Keeling *et al.*, 2008] from continuous atmospheric sampling by the Scripps Institute of Oceanography with ice core data from Law Dome [Etheridge *et al.*, 1996; MacFarling Meure *et al.*, 2006] and Siple [Friedli *et al.*, 1986]. Uncertainties are set to 0.5 ppm for the monthly direct samplings, 2 ppm for Law Dome and 3 ppm for Siple.

[6] Emissions from fossil-fuel use and cement production are taken from Boden *et al.* [2009] and land use emissions from Houghton [2008]. For 2007, a 3.3% increase in fossil-fuel emissions is assumed [Netherlands Environmental Assessment Agency, 2009], while land use emissions after 2005 are kept constant for lack of newer data. Land use emissions are assigned uncorrelated uncertainties for annual values of 10% (estimated from the interannual variability of the data), and a systematic error of 60%, equal to the decadal uncertainty given by Prentice *et al.* [2001] and implemented as a scaling parameter over the entire analysis period (see below). No uncertainty has been assigned to the fossil-fuel emissions.

[7] The Niño-3 sea-surface temperature (SST) [NOAA, 2009] is used as a statistical indicator to reflect changes in El Niño/Southern Oscillation, as well as the Volcanic Aerosol Index (VAI) [Ammann *et al.*, 2003] for the presence of climate-relevant stratospheric aerosols. It has been observed that atmospheric CO₂ increases faster if Niño3-SST is above average and more slowly if VAI is high [Jones and Cox, 2005; Knorr *et al.*, 2007]. No uncertainties are assigned to those indices.

[8] The CO₂ concentration data with uncertainties are converted to atmospheric growth in GtC by matrix multiplication applying finite differences. The same matrix is then used to compute the error covariance matrix of the growth rate. For the directly sampled data, the growth rate is

¹Department of Earth Sciences, University of Bristol, Bristol, UK.

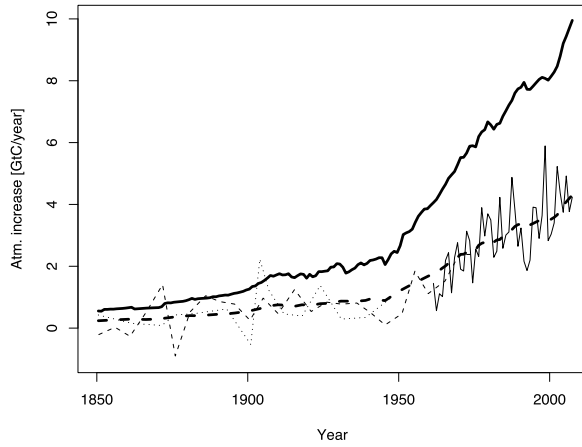


Figure 1. Observed atmospheric CO₂ increase derived from direct measurements, taking the average of Mauna Loa (Hawaii) and the South Pole (thin solid line), and two ice cores: Law Dome (dashed thin line) and Siple (dotted thin line). This is compared to total anthropogenic emissions (thick solid line) and 46% of total emissions (thick dashed line).

calculated after computing annual means from partially gap-filled data, but considering the number of valid monthly data entries for error propagation. Annual means are taken from July to the following June, so that growth rates are centered at mid-year. The result is a continuous time series of annual CO₂ increase from 1961 to 2007. For Law Dome, means and uncertainties of CO₂ concentrations are converted to five-year averages before calculating finite differences. No averaging is done for the data from Siple station.

3. Methods

[9] The basic statistical model into which the data are assimilated describes R , the rate of growth of atmospheric CO₂, as a function of a time varying airborne fraction ($f_0 + s \times t$) times fossil-fuel (F) plus land use (L) emissions plus two linear factors depending on Niño-3 SST anomalies (N) and volcanic aerosol index (V); t is defined as calendar year minus 2000:

$$R(t) = (f + st)[F(t) + lL(t)] + nN(t) + vV(t) + \varepsilon(t) \quad (1)$$

ε is the residual error and l a scaling parameter that describes the systematic uncertainty of the land use emissions. ε is described by a Gaussian distribution and includes both data and model uncertainty. The prior value of l is 1 and the prior uncertainty $\sigma_l = 0.6$ (see below). V is annual VAI, and N Niño-3 SST anomaly with a 4-month lag between concentrations and SSTs (SSTs leading) and then annually averaged. The time lag was found to produce the highest correlation in a lagged-correlation analysis of V and N against time de-trended atmospheric growth rate ($r = 0.79$). The SST anomaly is computed by subtracting the 1961–2007 mean from the annual values. n and v are further parameters besides f , s and l . Prior to 1961, where no direct measurements enter the model, V and N are set to 0. An additional set of de-trended indicators for volcanic aerosols and ENSO are computed by a linear least squares fit through the annual data and retaining the residual as N_d and V_d , respectively.

[10] The parameter estimation follows standard least-squares procedures. It is performed by minimising a cost function J defined as:

$$J = \frac{1}{2}(\mathbf{R} - \mathbf{D})^T \mathbf{C}^{-1} (\mathbf{R} - \mathbf{D}) + \frac{1}{2} \frac{(l - 1)^2}{\sigma_l^2}, \quad (2)$$

where \mathbf{R} is the rate of growth written as a vector over time steps, i.e., $\mathbf{R} = \{R(t_1), R(t_2), \dots, R(t_m)\}$ where t_1 to t_m are the times for which atmospheric CO₂ growth data is available, $\mathbf{D} = \{D(t_1), D(t_2), \dots, D(t_m)\}$ the vector of those atmospheric data ($D(t)$ is observed atmospheric growth rate), \mathbf{C} the error covariance matrix of the data and model and $\sigma_l = 0.6$. σ_l represents the systematic uncertainty of the emissions that scale over the entire time series and is consistent with the ca. 60% uncertainty of the decadal total land use emissions given by *Prentice et al.* [2001].

[11] \mathbf{C} is a $m \times m$ matrix again over the m points of time where data is available. It is the error covariance matrix for the difference between data and model, ε , and thus contains both data and model error [Tarantola, 1987; Rayner et al., 2005]. It is computed as

$$\mathbf{C} = \mathbf{C}_D + \mathbf{C}_s + \mathbf{C}_L. \quad (3)$$

[12] \mathbf{C}_D is the error covariance matrix of the atmospheric growth data, \mathbf{C}_s is the error covariance matrix of the statistical model, and \mathbf{C}_L the error arising from the uncorrelated uncertainties of L .

[13] Two sets of optimizations of the parameters of equation (1) are performed: in the first set, \mathbf{C}_s and \mathbf{C}_L are 0 and $\sigma_l \rightarrow \infty$, so no data error enters the estimation of the parameters. For the second set, the statistical model error is estimated from a previous optimization out of the first set via

$$\mathbf{C}_s = \frac{1}{d} \sum_{i=1}^n \varepsilon_0(t_i)^2. \quad (4)$$

[14] ε_0 is the residual of the previous optimization, d is degrees of freedom and $\mathbf{C}_s = C_s \mathbf{U}$, where \mathbf{U} is the unity matrix. The additional model error arising from the uncorrelated error of the land use emissions, \mathbf{C}_L , is estimated from

$$C_{L,i} = [\sigma_L(f_0 + s_0 t_i)L(t_i)]^2. \quad (5)$$

where f_0 and s_0 are again parameters f and s taken from the previous optimization without data uncertainties. $C_{L,i}$ are the diagonal elements of \mathbf{C}_L , with off-diagonal elements equal to 0.

4. Results

[15] The simplest model of the atmospheric growth rate is one of a constant AF and yields $f = 0.43$ when fitted to all data. How well this simple model reproduces the observations at the multi-decadal time scale is shown in Figure 1. Interannual fluctuations are seen in the direct measurements. The difference between the two ice core records gives an impression of the likely uncertainty.

[16] Seven further optimizations are performed and the results shown in Table 1. Versions 1 and 2 neglect data

Table 1. Results of the Fit of the Statistical Model to Data of Atmospheric CO₂ Growth With and Without Inclusion of Data Uncertainty and Other Factors^a

Version	Data Uncertainty	Ice Core Data	N, V Time De-trended	f	s [yr ⁻¹]	l	n [GtC yr ⁻¹ K ⁻¹]	v [GtC yr ⁻¹]
1	no	no	no	0.450±0.022	0.0014±0.0014 ^b	1 ^c	0 ^c	0 ^c
2	no	no	no	0.453±0.014	-0.0013±0.0009 ^d	1 ^c	0.98±0.12	-15±2
3	yes ^e	no	no	0.518±0.064	-0.0002±0.0017 ^b	0.25±0.60	0 ^c	0 ^c
4	yes ^f	no	no	0.468±0.047	-0.0018±0.0014 ^b	0.82±0.54	0.98±0.14	-15±3
5	yes ^e	yes	no	0.468±0.051	0.0007±0.0014 ^b	0.73±0.54	0 ^c	0 ^c
6	yes ^f	yes	no	0.514±0.035	-0.0019±0.0016 ^b	0.36±0.35	0.91±0.13	-14±2
7	no	no	yes	0.449±0.014	0.0012±0.0009 ^g	1 ^c	0.98±0.13	-15±3

^a f is airborne fraction in 2000, s is the trend in the airborne fraction, l a scalar for land use emissions, and n and v parameters describing the impact of ENSO (N) and volcanic aerosols (V).

^bNot significant.

^cFixed value.

^dSignificant at 85%.

^eModel uncertainty based on Version 1.

^fModel uncertainty based on Version 2.

^gSignificant at 82%.

uncertainties ($C = C_s, \sigma_l \rightarrow \infty, l$ fixed) and are only applied to direct CO₂ measurements. Versions 3 and 4 additionally include data uncertainties, and the last two but one also data from the ice core record. Version 7 is included for compatibility with the results by *Canadell et al.* [2007] and replaces $N(t)$ and $V(t)$ by $N_d(t)$ and $V_d(t)$. In each of the groups of two, the first has n and v fixed at 0 (no predictors of interannual variability), while the second includes optimization of n and v . Remember that f represents the airborne fraction in 2000.

[17] The most important result is that inclusion of data uncertainties moderately increases the uncertainty of the trend estimate, s . Furthermore, the use of the interannual predictors ($nN + vV$) hardly reduces the uncertainty in s . The trend itself is either very close to zero (Versions 3 and 5), or slightly negative when using interannual predictors (Versions 4 and 6). In none of the cases there is a significant trend.

[18] The posterior parameter l is always found to have a mean less than one, but with a large uncertainty. This indicates that the linear model of the AF fits the observations better if the land use emissions are scaled down, but that the absolute level of land use emissions remains highly uncertain. This impacts on AF and its level of uncertainty: if the posterior mean of l is low (Versions 3 and 6), the posterior mean of f is higher because lower emissions mean a larger fraction of CO₂ stays in the atmosphere.

[19] Figure 2 offers an explanation for why l is reduced. The predicted emissions of Versions 3 and 4 (blue and green dashed lines) are mostly lower than the unadjusted emissions (black dashed), but increase more rapidly in relative terms after about 2000. In other words, an apparent recent increase in the AF can also be reproduced by downscaling the land use emissions, which is the solution preferred by the optimization compared to increasing s . Figure 2 also shows how well Version 4 (solid green line) reproduces the interannual variability seen in the observations [cf. *Jones and Cox, 2005; Knorr et al., 2007*].

[20] Version 7 shows s to be positive at a 82% significance level (Table 1). This result is comparable to the one by *Canadell et al.* [2007], who also used a de-trended ENSO index (using the Southern Oscillation Index instead of SSTs) and VAI. If de-trending is abandoned, however, a

negative trend emerges (Version 2). This behavior is a result of trends in N (0.0082 Kyr⁻¹) and V (-0.00043 yr⁻¹). With the optimal values of n and v (Version 2), these translate to a predicted total trend of 0.0145 GtCyr⁻². Including this additional predicted trend requires $s < 0$ to match the observations, unless trends in Niño-3 SST and VAI have deliberately been adjusted to zero.

5. Discussion

[21] The present analysis has identified some ambiguity regarding the inclusion of interannual predictors into the statistical model used to detect a trend in the AF. Whether it is advisable to do so or not depends on the question asked. If we want to detect a trend caused by climate change, then the same climate change may be responsible

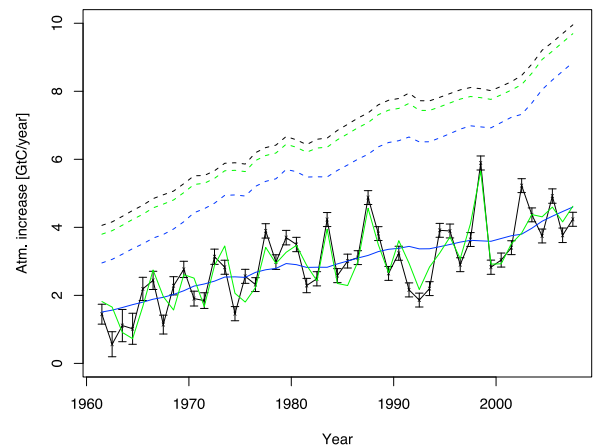


Figure 2. Observed (solid black line, with error bars) and predicted (solid lines) interannual variations of the atmospheric CO₂ growth rate compared to assumed total anthropogenic emissions (dashed lines). Green: optimization including statistical predictors of interannual variability. Blue: optimization without those predictors. Black dashed line: Unscaled emission estimate. The optimized emissions estimates scale land use emissions down to 82 and 25% of the original estimate, respectively.

for the observed trend in the ENSO signal. De-trending N then prevents the trend in the ENSO signal to be subtracted from s . However, the negative trend in the VAI is unrelated to the expected causes of a change in the AF. It would by itself be expected to lead to additional CO₂ remaining in the atmosphere ($\nu < 0$, Table 1), i.e. an increase in the AF. This additional trend should be subtracted from the observed AF trend if the goal is to detect the underlying long-term behavior. In any case, inclusion of ENSO and VAI has not led to reduced uncertainties of the trend parameter, s .

[22] Another finding is that reducing the land use emissions by a scalar causes total emissions to be more consistent with a model of a constant airborne fraction. It is difficult to rate this as a strong indication that land use emissions are systematically overestimated, as it depends on the validity of the statistical model. The possibility, however, presents itself and, given the evidence from oxygen data [Bopp *et al.*, 2002], would mean that a larger proportion of emissions is taken up by the ocean than what has been previously assumed. The analysis also shows that recent trends after 2000 can be explained by re-scaling land use emissions within their uncertainty ranges.

[23] The present treatment of systematic uncertainties in the land use emissions is rather simple, as systematic errors might have changed over time. Little is known about this, but inclusion of more parameters to allow temporal changes in systematic errors would have increased the uncertainty of the trend estimate. Uncertainties for fossil-fuel emissions, which are relatively small, have also been neglected. The uncertainty bounds reported here have therefore to be considered optimistic and, in the context of the question, conservative.

[24] Without the inclusion of ENSO and VAI in the analysis, the trend derived with data uncertainties is found to be very small, only 0.7 ± 1.4 or $0.2 \pm 1.7\%$ per decade, depending on whether the ice core record has been included or not. This is not significantly different from zero and in contrast to the previously published result [Canadell *et al.*, 2007] reporting an increase of $2.5 \pm 2.1\%$ per decade, but obtained with de-trended VAI and ENSO index and without accounting for data uncertainties. The equivalent result reported here is $1.2 \pm 0.9\%$ per decade. The difference between the last two probably reflects remaining differences in the method chosen.

6. Conclusion

[25] From what we understand about the underlying processes, uptake of atmospheric CO₂ should react not to a change in emissions, but to a change in concentrations. A further analysis of the likely contributing processes is necessary in order to establish the reasons for a near-constant AF since the start of industrialization. The hypothesis of a recent or secular trend in the AF cannot be supported on the basis of the available data and its accuracy.

[26] Given the importance of the AF for the degree of future climate change, the question is how to best predict its future course. One pre-requisite is that we gain a thorough understanding of why it has stayed approximately constant in the past, another that we improve our ability to detect if and when it changes. The most urgent need seems to exist for

more accurate estimates of land use emissions. Another possible approach is to add more data through the combination of many detailed regional studies such as the ones by Schuster and Watson [2007] and Le Quéré *et al.* [2007], or using process based models combined with data assimilation approaches [Rayner *et al.*, 2005]. If process models are used, however, they need to be carefully constructed in order to answer the question of why the AF has remained constant and not shown more pronounced decadal-scale fluctuations or a stronger secular trend.

[27] **Acknowledgments.** The author wishes to thank Renato Spahni for help with interpreting the ice core records, Thomas Kaminski for advice with data assimilation, Colin Prentice for inspiring discussions and Ian Enting and one anonymous reviewer for helpful comments. This research was funded by the U.K. Natural Environment Research Council under its QUEST program.

References

- Ammann, C. M., G. A. Meehl, W. M. Washington, and C. S. Zender (2003), A monthly and latitudinally varying volcanic forcing dataset in simulations of 20th century climate, *Geophys. Res. Lett.*, *30*(12), 1657, doi:10.1029/2003GL016875.
- Boden, T. A., G. Marland, and R. J. Andres (2009), Global CO₂ emissions from fossil-fuel burning, cement manufacture, and gas flaring: 1751–2006, Carbon Dioxide Inf. Anal. Cent., Oak Ridge Natl. Lab., Oak Ridge, Tenn.
- Bopp, L., C. Le Quéré, M. Heimann, and A. C. Manning (2002), Climate-induced oceanic oxygen fluxes: Implications for the contemporary carbon budget, *Global Biogeochem. Cycles*, *16*(2), 1022, doi:10.1029/2001GB001445.
- Canadell, J. G., C. Le Quéré, M. R. Raupach, C. B. Field, E. Buitenhuis, P. Ciais, T. J. Conway, N. P. Gilett, J. T. Houghton, and G. Marland (2007), Contributions to accelerating atmospheric CO₂ growth from economic activity, carbon intensity, and efficiency of natural sinks, *Proc. Natl. Acad. Sci. U. S. A.*, *104*, 18,866–18,870, doi:10.1073/pnas.0702737104.
- Etheridge, D. M., L. P. Steele, R. L. Langenfelds, R. J. Francey, J.-M. Barnola, and V. I. Morgan (1996), Natural and anthropogenic changes in atmospheric CO₂ over the last 1000 years from air in Antarctic ice and firn, *J. Geophys. Res.*, *101*, 4115–4128, doi:10.1029/95JD03410.
- Friedli, H., H. Löttscher, H. Oeschger, U. Siegenthaler, and B. Stauffer (1986), Ice core record of ¹³C/¹²C ratio of atmospheric CO₂ in the past two centuries, *Nature*, *324*, 237–238, doi:10.1038/324237a0.
- Friedlingstein, P., et al. (2006), Climate–carbon cycle feedback analysis, results from the C4MIP model intercomparison, *J. Clim.*, *19*, 3337–3353, doi:10.1175/JCLI3800.1.
- Houghton, R. A. (2008), Carbon flux to the atmosphere from land-use changes, in *Trends: A Compendium of Data on Global Change*, Carbon Dioxide Inf. Anal. Cent., Oak Ridge Natl. Lab., Oak Ridge, Tenn.
- Jones, C. D., and P. M. Cox (2005), On the significance of atmospheric CO₂ growth rate anomalies in 2002–2003, *Geophys. Res. Lett.*, *32*, L14816, doi:10.1029/2005GL023027.
- Keeling, R. F., S. C. Piper, A. F. Bollenbacher, and J. S. Walker (2008), Atmospheric CO₂ records from sites in the SIO air sampling network, in *Trends: A Compendium of Data on Global Change*, Carbon Dioxide Inf. Anal. Cent., Oak Ridge Natl. Lab., Oak Ridge, Tenn.
- Keeling, R. F., S. C. Piper, A. F. Bollenbacher, and J. S. Walker (2009), Atmospheric CO₂ records from sites in the SIO air sampling network, in *Trends: A Compendium of Data on Global Change*, Carbon Dioxide Inf. Anal. Cent., Oak Ridge Natl. Lab., Oak Ridge, Tenn.
- Knorr, W., N. Gobron, M. Scholze, T. Kaminski, R. Schnur, and B. Pinty (2007), Impact of terrestrial biosphere carbon exchanges on the anomalous CO₂ increase in 2002–2003, *Geophys. Res. Lett.*, *34*, L09703, doi:10.1029/2006GL029019.
- Le Quéré, C., et al. (2007), Saturation of the Southern Ocean CO₂ sink due to recent climate change, *Science*, *316*, 1735–1738, doi:10.1126/science.1136188.
- MacFarling Meure, C., D. Etheridge, C. Trudinger, P. Steele, R. Langenfelds, T. van Ommen, A. Smith, and J. Elkins (2006), Law Dome CO₂, CH₄ and N₂O ice core records extended to 2000 years BP, *Geophys. Res. Lett.*, *33*, L14810, doi:10.1029/2006GL026152.
- Netherlands Environmental Assessment Agency (2009), Global CO₂ emissions: annual increase halves in 2008, PBL report, Neth. Environ. Assess. Agency, Bilthoven, Netherlands. (Available at <http://www.pbl.nl/en/publications/2009/Global-CO2-emissions-annual-increase-halves-in-2008.html>)

- NOAA (2009), Monthly Niño-3 sea surface temperature, <http://www.cpc.noaa.gov/data/indices/sstoi.indices>, Clim. Predict. Cent., Camp Springs, Md.
- Prentice, I. C., et al. (2001), The carbon cycle and atmospheric carbon dioxide, in *Climate Change 2001: The Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change*, edited by J. T. Houghton et al., pp. 185–225, Cambridge Univ. Press, Cambridge, U. K.
- Raupach, M. R., J. G. Canadell, and C. Le Quéré (2008), Anthropogenic and biophysical contributions to increasing CO₂ growth rate and airborne fraction, *Biogeosciences*, *5*, 1601–1613.
- Rayner, P. J., M. Scholze, W. Knorr, T. Kaminski, R. Giering, and H. Widmann (2005), Two decades of terrestrial carbon fluxes from a carbon cycle data assimilation system (CCDAS), *Global Biogeochem. Cycles*, *19*, GB2026, doi:10.1029/2004GB002254.
- Schuster, U., and A. J. Watson (2007), A variable and decreasing sink for atmospheric CO₂ in the North Atlantic, *J. Geophys. Res.*, *112*, C11006, doi:10.1029/2006JC003941.
- Tarantola, A. (1987), *Inverse Problem Theory: Methods for Data Fitting and Parameter Estimation*, Elsevier, New York.
- Trenberth, K. (1981), Seasonal variations in global sea level pressure and the total mass of the atmosphere, *J. Geophys. Res.*, *86*, 5238–5246, doi:10.1029/JC086iC06p05238.

W. Knorr, Department of Earth Sciences, University of Bristol, Wills Memorial Bldg., Queen's Rd., Bristol BS8 1RJ, UK. (wolfgang.knorr@bristol.ac.uk)