Differences between trends in atmospheric CO$_2$ and the reported trends in anthropogenic CO$_2$ emissions

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ABSTRACT
Averaged annual accumulation of CO$_2$ in the atmosphere, $\frac{dC}{dt}$, has been slowing from peak growth in 2002/2003 associated with anomalous climate-induced emissions at high northern latitudes. This slowing is widespread but determined with greatest certainty in the largest well-mixed portion of the global troposphere ($30^\circ$S–$90^\circ$S). We rely on atmospheric mixing for global integration and selection of atmospheric data for spatial representativeness. Prior to 2002/2003, after empirical adjustment for perturbations associated with ENSO and volcanic activity (EV), $\frac{dC}{dt}$ increases are well represented by linear regression, using direct monitoring records from 1990 or 1965, also from pre-industrial times using archived air. In contrast, modelled atmospheric trends due to reported emissions $\frac{dC}{dt}$ (assuming historically consistent oceanic and terrestrial uptake mechanisms), agree with $\frac{dC}{dt}$ up until 1990, are near-stable through the 1990s and increase by 29% between 2000 and 2008. Using atmospheric constraints based on trends in both $\frac{dC}{dt}$ and interhemispheric gradient, the differences between trends in $\frac{dC}{dt}$ and atmospheric CO$_2$ growth are most simply explained as an artefact of underestimating 1994–2003 emissions by around 6%. This is achieved with a near constant post-1965 airborne fraction; otherwise unusually complicated sink changes are required for the period.

1. Introduction
Canadell et al. (2007) conclude that 2000–2006 economic activity, carbon intensity and efficiency of natural sinks contribute to accelerating atmospheric CO$_2$ growth. Expanded detail is provided by Raupach et al. (2007, 2008). They include an estimate of ocean uptake by Le Quéré et al. (2007), who report reduced effectiveness of the Southern Ocean CO$_2$ sink due to climate change. These studies use emission estimates of fossil fuel burning and cement-production by Marland et al. (2008), and land-use change (LUC) by Houghton (2008), based largely on national emissions reported to the United Nations Framework Convention on Climate Change (UNFCCC). Recent increasing emissions are attributed primarily to Asia (Gregg et al., 2008). More recently, Le Quéré et al. (2009) highlight a 29% increase in reported emissions since 2000 and conclude that reduced uncertainty in natural sinks is required to improve definition of the global carbon cycle. A decreasing trend in natural sinks is supported by modelled global changes in ocean uptake (Sarmiento et al., 2009) and, for example, by changes in South Indian Ocean fugacity measurements relative to that in the atmosphere (Metzl, 2009). In the Le Quéré and Sarmiento inversion studies, atmospheric CO$_2$ growth is derived from concentrations reported to international databases, with little discussion of potential measurement or sampling bias.

Our focus has been minimization of uncertainty in global trends in atmospheric CO$_2$ growth ($\frac{dC}{dt}$), which, irrespective of contributing processes, relate most directly to changes in climate forcing.

After a general discussion of measurement and sampling bias in Section 2, Section 3 demonstrates an overall slowing in $\frac{dC}{dt}$ since 2002/2003 in a record from Cape Grim (Tasmania, 41°S) employing in situ CO$_2$ monitoring with unprecedented temporal precision and, more significantly, rigorous selection of hourly data that results in unusually large spatial representativeness.

The growth trend is assessed against historic data introduced in Section 4, which includes four-decade trends in Scripps Institution of Oceanography records (Keeling et al. 2001, 2005; http://scrippsco2.ucsd.edu/data/data.html) from Mauna Loa (MLO Hawaii, 21°N) and/or South Pole (SPO, 90°S). We also use the trend in interhemispheric gradient in mixing ratio (MLO–SPO). On subdecadal timeframes, the four-decade Scripps records are used to provide an empirical description relating interannual variations in $\frac{dC}{dt}$ with climate and volcanic
activity, permitting assessment of their likely contribution to the
most recent Cape Grim \( \frac{dC_t}{dt} \) trends. Section 4 concludes with
an estimate of the atmospheric trends (\( dC_t/dt \)) anticipated from
reported emissions, which are obtained using a simple box model
that closely describes the carbon budget on decadal timescales
since pre-industrial times.

Section 5 extends the analysis of CSIRO CO\(_2\) measurements
over two decades and provides additional constraints on the
location of emissions and sinks by way of relative trends at
different latitudes, using data from a global flask sampling
network (these are more representative of data submitted to
global databases, for example, http://gaw.kishou.go.jp/wdcgg/;
http://www.esrl.noaa.gov/.../globalview). The flask \( dC_t/dt \) data
are processed in a similar manner to that used in Section 3,
and growth rate uncertainties compared. The box model is used
to estimate the magnitude of source and sink changes that are
required to reconcile the differences in the recent \( dC_t/dt \) and
\( dC_t/dt \) trends.

The analyses are discussed in Section 6 and conclusions sum-
marized in Section 7.

2. Sampling and selection issues

2.1. Site selection

Optimal sampling strategies to obtain global CO\(_2\) growth rate
changes are different from those that seek information on
regional carbon budgets. Averaging data from many sites, to
address sampling and measurement uncertainties (WMO, 2008;
http://www.esrl.noaa.gov/gmd/ccgg/trends/index.html#global),
incorporates sampling bias and can obscure fine detail in high-
quality records. This is a critical issue for Northern Hemispher-
(NH) boundary-layer sampling sites that are embedded in an
atmospheric mixing volume overlaying 70% of the global land
mass, associated with the bulk of industrial emissions and
also includes major rapidly exchanging, climate-sensitive plant
carbon reservoirs. Even with common hemispheric climate
forcing and relatively rapid within-hemisphere mixing (of order
3 weeks), large spatial (horizontal and vertical) and temporal
heterogeneity in NH CO\(_2\) exists.

Denning et al. (2004) report that interhemispheric CO\(_2\) mixing
times for 12 atmospheric transport models range from 0.5
to 1.3 yr for 3-D models (longer for 1-D and 2-D models).
The main barrier to N–S mixing is at the southern edge of the
interropical convergence zone (Miyazaki et al., 2008). Since
the time for interhemispheric mixing (is much greater than
that for within-hemisphere mixing, air sampled south of 30 \(^\circ\)S
provides an effective integration of the heterogeneous northern
fluxes, limiting the possibility of overrepresentation of re-
gional northern fluxes in a global total. Then, selection of mid-
to-high latitude southern air to minimize recent land contact can
lead to significant advantages in representing global growth rate
changes.

This argument depends on the very small spatial gradients
and interannual variability in those gradients at mid-to-high
southern latitudes; for example, typical background CO\(_2\) fields
>30 \(^\circ\)S have annual horizontal gradients <0.2 ppm, vertical
gradients <0.1 ppm km\(^{-1}\), well-characterized seasonal ampli-
tude ~0.5 ppm. Aircraft vertical CO\(_2\)-profiling summarized by
Stephens et al. (2007), presents mid-day profiles from 12 global
locations. The Cape Grim (41 \(^\circ\)S) data, representing 10 yr of
monthly flights in clean air conditions, show the smallest gradi-
ents, seasonally and vertically, and are given emphasis below. Of
the NH sites, the Hawaiian site Mauna Loa best exhibits these
properties and is treated independently in this study as a way of
indicating sensitivity to hemisphere differences.

2.2. Measurement error

There are several potential growth rate errors in CO\(_2\) time series
that are not well addressed in international data repositories or
metadata. They are examined in detail in Appendix S2, estab-
lishing that resulting bias in the CSIRO records used here is
small, and more generally, errors are less likely to be a signifi-
cantly influence after the 1980s. Thus, averaging data from many
sites to overcome measurement error is unjustified, particularly
when larger uncertainty due to sampling bias can be introduced
by the averaging process.

2.3. Data selection

Selecting data from individual site records to reconstruct global
trends requires attention to potential bias in measured growth
rate, and to sampling strategies that minimize local or regional
contamination. Cape Grim provides unusual opportunities to
assess both issues by having several CSIRO operated CO\(_2\) mon-
itoring programs with different measurement, calibration and
sampling strategies. For example, they include an on-going au-
tomated comparison of measurements on the same flask-air
that closely compares CSIRO and National Oceanic and At-
mospheric Administration USA, Earth System Research Labo-
atory (NOAA ESRL) measurements at this site (Masarie et al.,
2001); differences in smoothed data from CSIRO and NOAA
programs remain within ~0.05 ppm.

We select ‘baseline’ CO\(_2\) hours that require moderate to high
wind speeds (>5 m s\(^{-1}\)) from the marine wind-direction sector
that meet a CO\(_2\) stability criterion of <0.3 ppm in the hourly
mixing ratios over five successive hours. For these hours, hourly
radon measurements (a signal imposed by air passage over land,
decaying with 3.8-d half life) confirm a marine boundary layer
origin. For example, from 2001 to 2007, 36% of hours with wind
speeds >2 m s\(^{-1}\) have a median concentration of 37 mBq m\(^{-3}\)
(Zahorowski et al., 2005; W. Zahorowski, personal communica-
tion 2009), compared to a 100 mBq m\(^{-3}\) we have historically
used as a baseline threshold. Appendix S1 shows typical 10-
d back trajectories for conditions when Cape Grim radon is
3. Growth rates from the Cape Grim LoFlo in situ monitoring

Since 2001, CO₂ has been monitored at Cape Grim, a location with frequent exposure to Southern Ocean marine boundary layer air, with unprecedented temporal stability, and with advanced infrastructure that enables selection and verification of the origin of the air. Of records examined, this provides recent multiyear trends in atmospheric CO₂ growth with the smallest uncertainty, and is used here to introduce methods to quantify the uncertainty.

The LoFlo analyser (Da Costa and Steele, 1999; Francey and Steele, 2003) demonstrates in monthly measurements of air in seven attached high pressure cylinders, each with mixing ratios assigned on the WMO Mole Fraction scale by the WMO Central CO₂ Calibration Laboratory; between 2001 and 2010 the standard deviation of monthly determinations of near-ambient cylinder air relative to the other six, and in more-transient ambient ‘reference’ gases relative to the seven-cylinder scale, is typically ±0.003 ppm. The analyser samples 190 ± 77 baseline hours per month (with a seasonal range of 135 ± 58 in April, 19% of hours, to 253 ± 44 in December, 34% of hours).

Figure 1a shows the mean monthly mixing ratio, and its uncertainty SD₁₂ calculated by combining the standard deviation (SD₁) of daily values subtracted from a smoothing spline with 50% attenuation at 80 d (mainly representing bias from incomplete sampling of the seasonal cycle throughout the month), and the standard deviation (SD₂) of the residuals of each selected hourly value about the smoothing spline for that month. Since 2001, the mean LoFlo monthly SD₁₂ = (SD₁² + SD₂²)½ is 0.15 ± 0.03 ppm.

Figure 1b derives growth rates, dCᵦ/dt, from the monthly mixing ratios using three different methods for calculating dCᵦ/dt. The smooth solid line represents the widely used time-differential of a 22-month smoothed trend curve through deseasonalized data (Thoning et al., 1989). High time resolution (1 mnth) growth rates are obtained by annual differencing of values for corresponding months, plotted at the mean time, and using the square root of the sum of squares. The 5-yr smoothed growth trends (5 yr), shown as a ±<u> shaded band, are obtained from the first derivative of a spline fit to the monthly mixing ratio data with 50% attenuation at 5 yr while propagating the SD₁₂ monthly uncertainties through the splines (Enting et al., 2006).
There is a high degree of consistency between the three methods of determining $\frac{dC_a}{dt}$. The 5-yr trend decreases from a peak of $2.11 \pm 0.02$ ppm yr$^{-1}$ in 2002.6, through $1.84 \pm 0.02$ ppm yr$^{-1}$ in 2008.5, to $1.79 \pm 0.04$ ppm yr$^{-1}$ in 2010.0. This is unexpected at a time when reported emissions from 2000 to 2008.5 have increased by 29%. Figure 1b also shows that the LoFlo trend is slowing relative to, and falling significantly below, the four-decade trend at SPO over this period (dashed linear regression, denoted ‘40 yr’). That this is unlikely to be the result of climate variation is implied by the reasonable agreement in the amplitude and phasing of interannual variability driven primarily by ENSO data (dashed curve). These dashed ‘baselines’ are derived in the next section (and used above with an initial assumption that monthly data can be scaled using empirical coefficients derived from 9-month smoothed $\frac{dC_a}{dt}$, and ENSO and volcanic indices).

4. Historic growth rate behaviour

Figure 2a shows the 22-month smoothed $\frac{dC_a}{dt}$ (Thoning et al., 1989) over five decades from MLO and SPO, where the large time span reduces the significance of measurement and sampling bias on mean behaviour. Data shown in Fig. 2 contribute to the following background information used in this study:

4.1. Linear $\frac{dC_a}{dt}$ trends

Figure 2a provides linear regressions through data from 1965 to 2006; the SPO regression is that used in Fig. 1b. Data prior to 1965 are not used due to significant data gaps in the SPO record (which may contribute to the unusual early interannual behaviour in the SPO $\frac{dC_a}{dt}$), and to the occurrence at the start of the record of explosive volcanic activity (Mt. Agung, 1963) that may unduly bias the regressions (in the sense of making the difference from the Fig. 1b LoFlo trend even greater). The fact that the MLO increase is marginally faster than that at SPO is consistent with increasing emissions that are mainly located in the NH.

4.2. Interhemispheric gradient

Figure 2b shows the difference ($C_{MLO} - C_{SPO}$) obtained from the 22-month smoothing spline through monthly mixing ratio data. It is well represented by the linear regression and is also attributed to increasing emissions that are mainly located in the NH. In Section 5.1, we use this relationship between $\frac{dC_a}{dt}$ and ($C_{MLO} - C_{SPO}$) to constrain the location of sources and/or sinks that might reconcile reported emission and measured atmospheric trends.

4.3. Interannual variability

The $\frac{dC_a}{dt}$ variation attributable to ENSO and volcanoes (EV) used in Fig. 1b is empirically estimated from the CO2 monthly data used in Fig. 2a, using an independently developed approach similar to that of Raupach et al. (2008).

Compared to the Raupach et al. (2008) approach, our analyses are restricted to data from 1965, after which we place significance in the fact that the amplitude of interannual variation at SPO is consistently equal to or less than that at MLO. Also, the ENSO and volcano time-series are extended, and weighted separately for positive (+) and negative (−) climate anomalies to accommodate the suggestion (Reichstein et al., 2009) that ‘the response of the ecosystem carbon balance to climate variability

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**Fig. 2.** (a) Long-term growth rates (ppm yr$^{-1}$) from Mauna Loa (MLO, 20°N) and South Pole (SPO, 90°S) measured by Scripps Institution of Oceanography. Growth rates are obtained by differentiation of the smoothed trend curve through monthly mixing ratios (Thoning et al., 1989). Linear regressions are fitted to the 1965–2006 data of each trend curve (and extrapolated to provide a baseline). Anomalous events thought to influence CO2 growth are indicated. The thick line with overlaid white symbols shows an estimate of changes due to reported anthropogenic emissions (Section 4.4). (b) The interhemispheric gradient obtained by subtracting the SPO mixing ratio trend from that at MLO, also fitted with a linear regression.
is highly skewed, i.e. large carbon losses are much more likely than large carbon gains during periods with climatic conditions deviating from normal’. Thus,

\[ EV = a_0 + a_1 \text{ENSO}^+(t - t_{\text{lag}}) + a_2 \text{ENSO}^-(t - t_{\text{lag}}) + a_3 \text{VOLC}(t), \]

where EV is obtained using least squares fitting of MLO or SPO growth rate anomalies, ENSO is a climate anomaly index that precedes a CO₂ growth variation by \( t_{\text{lag}} \) months (\( t_{\text{lag}} \) ranges between 3 and 12 months); VOLC describes the influence of major volcanic eruptions. Using 1965–2006 MLO and SPO data with Fig. 2 regressions subtracted, we solve for \( a_i \) and \( t_{\text{lag}} \) using different ENSO and VOLC anomalies (selected examples shown in Figs 3a, 3b. Appendix S3 provides correlation \( r \) and lag details for a range of ENSO and volcanic indices, and smoothing parameters of 5, 9 and 22 months. For all indices, the EV are derived separately for MLO and SPO \( \frac{dC}{dt} \).

Selecting the 9-month smoothing on all parameters, Figs 3c and d compare EV \( (r = 0.8, t_{\text{lag}} = 4 \text{ months}) \) with the MLO, SPO \( \frac{dC}{dt} \) anomalies. Only results for Niño3.4 (sea-surface temperature anomalies generated to highlight climate sensitivity) are presented here for discussion. The EV explain up to 60% of the interannual variability in \( \frac{dC}{dt} \), and are insensitive to site or smoothing. The positive anomalies are marginally more strongly correlated; all tested indices provide similar overall conclusions, as discussed in Appendix S3.

Examination of Fig. 3 suggests that unexplained CO₂ growth-rate variation is mainly systematic rather than randomly distributed noise, and results from variations in the magnitude of responses to ENSO and volcanoes. Since, on average, the EV reflect CO₂ fluxes from equatorial vegetation (Rayner et al., 2008), it is likely that the proximity of individual equatorial flux events to particular measurement sites, or non-equatorial events, contribute much of the unexplained variance. If the processes that lead to CO₂ perturbations varies between ENSO events this may also contribute to variance.

With this perspective we consider separate interannual events in the context of available independent evidence for unusual flux processes or anomalous climate forcing. Information about these events can define the sense, if not the magnitude of anomalous contributions, and need to be considered when presenting statistically based conclusions. For example, differences in forcing processes are indicated for recent interannual events in 1992–1994, 1997–1998, 1999–2002 and 2002–2003:

- The 1992–1994 perturbation is widely attributed to volcanic activity (e.g. Jones and Cox, 2001).
- The 1997–1998 departures are associated with record ENSO emissions from the equatorial Indonesian wild fires.
involving unusual contribution from burning of recently exposed peat (Langenfelds et al., 2002; Page et al., 2002). It will be seen below that this signal is similarly expressed in northern and southern latitudes.

- The 1999–2002 dip is yet to be specifically addressed in the literature, though it does coincide with the major anomaly since 1982 in the δ13CO2 versus CO2 record (Allison and Francey, 2007). The timing also corresponds to unique anomalies in ocean records over the period (Ashok et al., 2007; Dore et al., 2009), also a quiet period in ENSO related wild fires (Randerson et al., 2005, 2009). This is the subject of a separate study.

- The 2002–2003 anomaly has been associated with drought in America and Europe (emissions of ~0.2–0.3 PgC; Vetter et al., 2008), and significant wild fire activity in Siberia (emissions of ~0.5 PgC; Rayner et al., 2008). It will be seen below that this event is expressed quite differently at northern and southern latitudes.

- We also note in passing, that interhemispheric difference anomalies in both CO2 and δ13CO2 suggest unusual northern terrestrial emissions in 2006.

As a further example, correlations summarized in Appendix S3 explore the influence of record length using post-1965 CO2 data, and post-1980 data only; for SPO data there is no difference, but MLO based correlations are stronger using post-1980 data, which we attribute to the regional proximity of MLO to the major equatorial events in the 1990s (probably aided by the mean location of the ITCZ being north of the equator).

4.4. Historic growth changes attributable to emissions

Figure 2a also includes a model estimate of the atmospheric growth trend that can be attributed to reported emissions. Trudinger et al. (1999) demonstrate close agreement over the last century between trends in reported emissions and smoothed CO2 growth (in contemporary post-1960 air plus Antarctic ice-core and bridging firn and archived air samples), using simple parametrizations of ocean and terrestrial sinks (see Appendix S4). Here we use updated estimates of emissions due to fossil fuel burning (from G. Marland) and land use change (from R. Houghton) used by Le Quéré et al. (2009). The modelled atmospheric growth changes attributable to these emissions, dC/E/dt, while not including natural interannual variability, provide a convenient baseline for examining recent observed dC/a/dt trends. Our modelled change in global sinks is an increase by ~0.9 PgC yr⁻¹ per decade (with partitioning not tightly determined but approximately equally distributed between land and oceans).

5. Measured atmospheric growth from global flask sampling

To get a global and two-decade perspective on the Fig. 1 Cape Grim LoFlo measurements discussed in Section 3, we use data from the CSIRO global atmospheric sampling network (Francey et al., 1996). However, it is instructive to first examine the relative trends between flask sites and flask measurement laboratories, using a simple method of comparing annual average mixing ratios to remove seasonal influence, and using differences from South Pole annual averages to suppress interannual variation that is similarly expressed in both hemispheres.

5.1. Relative trends

In Fig. 4 we present CO2 data from the CSIRO global flask network for which consistent two-decade measurements exist. Italicized lower case site labels are used for CSIRO flask records to distinguish them from Scripps MLO and SPO sites and measurement methodology, and include sampling from Alert (alc,
82°N), Mauna Loa (mlu, 20°N), Cape Ferguson (cfa, 19°S), Cape Grim (cga, 41°S) and Macquarie Island (mqa, 54°S).

The difference between Scripps and CSIRO increases slowly to ~0.1 ppm over two decades (most clearly seen in the SPO-spu record, however the mean difference MLO-mlu before 2000 is positive, and after 2000 is negative, the change being of similar sign and magnitude to the change in SPO-spu). Possible calibration bias in one or other (or both) of the laboratories is discussed in Appendix 2. The difference is not significant in the present context, particularly when we use only CSIRO data that is precisely linked to a common local scale. Temporal consistency in the differences is well within 0.1 ppm.

The very small measured spatial gradients and small interannual variability at the mid-to-high southern latitudes are consistent with the CO₂ homogeneity in this large globally representative region. The increasing relative trends in the mlu, alc and MLO records are consistent with NH emissions being primarily responsible for increasing CO₂, with larger interannual variability reflecting the greater NH land/transport influences. A linear regression through the mlu-spu data in Fig. 4 gives a similar slope of 0.057 ± 0.011 ppm yr⁻¹ compared to the long-term slope in Fig. 2b of 0.047 ± 0.001 ppm yr⁻¹. Interannual variability at cfa (on the Great Barrier Reef, 19°S) reflects proximity to land sources and more complex meteorology, and possibly reflects Indonesian peat fires in 1997/98 (Langenfelds et al., 2002; Page et al., 2002); however an overall trend in cfa relative to spu is not evident.

Note that a significant early 1990s volcanic anomaly is not evident in Fig. 4 despite the well-documented volcanic influence on global growth rates (Jones and Cox, 2001). The absence of a perturbation on the interhemispheric difference is consistent with CO₂ uptake by the equatorial terrestrial biosphere (as a result of environmental changes triggered by the Pinatubo eruption), a perturbation that has been propagated similarly to both hemispheres.

In Fig. 2, the consequence of NH fossil emissions over four decades is a near-linear 1 ppm yr⁻¹ change in dCₐ/dt, matched by a near-linear 2 ppm change in Cₐ/Cₐₚₑ, with both trends attributed to increasing anthropogenic emissions in the NH. The post-2000 dCₐ/dt increase of 0.7 ppm yr⁻¹ over 7 yr is predominantly attributed to NH Asian fossil emissions data (Gregg et al., 2008). In the absence of other explanations for these trends, we estimate (on time frames long compared to interhemispheric mixing) changes in N–S gradient on the basis of its historic relationship with emissions. Based on the relationship between the linear regression slopes in Figs 2a and b the Mauna Loa–South Pole difference expected from dCₐ/dt is illustrated by the dashed line superimposed on the mlu-spu curve of Fig. 4. The estimated 1990s flattening and ~1.4 ppm Cₐ=Cₐₚₑ change since 2000 are not observed in the flask interhemispheric gradient, with the biggest difference occurring in the decade starting in 1992. We rather observe a gradual increase since 1992.

5.2. Flask growth rates

We limit dCₐ/dt discussion to Mauna Loa (mlu) and Cape Grim (cga) flasks which, of data that we have detailed information, best represent their respective hemispheres. Figure 5 highlights uncertainties in growth-rates on both monthly and 5-yr smoothed time frames, for comparison with Fig. 1. Consistent with our suggestion that mlu is more susceptible to nearby equatorial and within-NH influences, the record exhibits larger scatter, and interannual excursions in the monthly data than cga, and a larger uncertainty band in the 5-yr trends. Of particular note is the 2002–2003 period which shows the largest consistent difference between interannual variability in mlu dCₐ/dt (symbols) and that in the EV index; this is far less evident at Cape Grim.

Here, monthly mixing ratio data are the mean of measurements on two to six flasks (<1 h total sampling in selected clean-air conditions) per month, whereas the LoFlo data of Fig. 1, despite much more rigorous selection criteria, involve 190 ± 77 h per month. Uncertainty combines SD1, the standard deviation of an 80-d smoothed curve through the individual flask data with the standard deviation SD2 of individual flask residuals around the smooth curve. Compared to the LoFlo record, where, in ppm, SD12 = 0.15 ± 0.03; for the flask records SD12 = 0.18 ± 0.07, 0.50 ± 0.22 for cga, mlu, respectively. The uncertainty for flask data at both sites is multiplied by 3 to recognize the sparse sampling of each month and less rigorous selection (for example, synoptic variability is not captured, and this is more serious at Mauna Loa due to the large seasonal and latitudinal gradients in CO₂ fields, discussed in Section 2.1). The 5-yr spline bands propagate ±3*SD12 uncertainties. However, the respective 5-yr trends agree within spline uncertainties and exhibit a similar pattern; the pattern is similar to that in the LoFlo record from 2001. Relative 5-yr trends (ppm yr⁻¹) in the three records are:

|        | Cape Grim (LoFlo) | Cape Grim (cga) | Mauna Loa (mlu) |
|--------|------------------|----------------|-----------------|
| 2002.6 | 2.11 ± 0.02      | 2.07 ± 0.13    | 2.20 ± 0.41     |
| 2008.5 | 1.84 ± 0.02      | 1.85 ± 0.16    | 1.68 ± 0.53     |

The close agreement between the Cape Grim LoFlo and cga trends suggests that the three times SD12 multiplier overestimates uncertainty at this site, and the less stringent selection criteria used for flask sample collection are effective in representing the large scale SH homogeneity. As calculated, the changes in the flask data are of marginal significance, even at mlu where the indicated decrease is largest.

5.3. Flask growth rate trends versus emission trends

To aid comparison of measured atmospheric trends to trends in emission estimates, we suppress ‘natural’ variability in the dCₐ/dt by subtracting the empirically derived EV (Section 4.3). Figure 6 includes the changes expected from the emission
Differences between Atmospheric CO₂ and Anthropogenic CO₂ Emissions

Fig. 5. The growth rate $dC_a/dt$ (ppm yr$^{-1}$) using monthly values with $\pm 3^*$SD12 uncertainties obtained from annual differences in the mean monthly mixing ratios; 5-yr smoothed values, shown with shading, represent the mean trend $\pm$ uncertainty obtained by differentiation of a smoothing spline (Enting et al., 2006). (a) Uses Mauna Loa ($mlu$) flask air collected and measured by CSIRO (open circles, with error bars plotted every third month). The MLO four-decade linear regression (light dashes), and regression plus EV$\text{nino3.4}$ (dark dashes), are included for comparison. (b) Uses Cape Grim ($cga$) flask air collected and measured by CSIRO (filled circles). The SPO four-decade linear regression (light dash), and regression plus EV$\text{nino3.4}$ (dark dash), are included for comparison. Panel (b) also includes the selected monthly LoFlo data from Fig. 1b (dots with error bars from 2002).

estimates, $dC_E/dt$, as a thick line with overlaid white symbols and the four-decade SPO $dC_a/dt$ as a linear, thin, dashed line.

In Fig. 6a, $cga$ and $mlu$ growth rates with uncertainties are obtained by applying the Enting et al. (2006) method on monthly mixing ratios $\pm 3^*$SD12 using 9-month smoothing; EV determined using 9-month smoothed monthly values of Niño3.4 anomalies, advanced by 4 months, are subtracted. Uncertainties combine the $dC_a/dt$ spline uncertainties with $0.4^*$Niño3.4 anomalies. Figure 6 shows ($dC_{mlu}/dt$-EV) and ($dC_{cga}/dt$-EV) with unadjusted $dC_{cga}/dt$ for comparison. The $mlu$ variability remains significantly greater than that at $cga$. The $cga$ cases show that adjustment for the major volcanic event in 1991–1994 is significant, but perhaps incomplete, since this is the most significant departure of ($dC_{cga}/dt$-EV) data from the four-decade SPO trend.

The 1997–1998 peak is reduced, and adjustment is noticeable around 2003. The small ENSO activity after 2003 is indicated by the small adjustments.

Overall, significant interannual variability persists in these curves (possibly due to reasons outlined in Section 4.3, and suggesting that a more sophisticated treatment of interannual variability may reduce the natural variability even further); however between 1994 and 2003 the $dC_a/dt$ passes through the lowest points in the ($dC_{cga}/dt$-EV) curve, rising to peak values in 2007. The EV adjustment does not provide an explanation for the $dC_{cga}/dt$ decline quantified in Fig. 1.

This is emphasized in Fig. 6b, with expanded y-scale, which uses 5-yr smoothing of the $dC_a/dt$-EV values. In this case, the uncertainties fed to the spline fitting are amplified to represent the persisting interannual variability seen in (a), using a minimum monthly uncertainty determined by the standard error in the y-residuals from a linear regression through the $dC_a/dt$-EV of Fig. 6a. This has the effect of reducing the difference between uncertainty bands from the three records. The $dC_{cga}/dt$ is well below these uncertainties between 1994 and 2003, rising above the uncertainty bands in 2007.
6. Discussion

From the early 1990s (after the large volcanic perturbation), Fig. 6 shows \(\frac{dC_e}{dt}\) systematically below ENSO adjusted atmospheric trends (\(\frac{dC_a}{dt}-EV\)) then rapidly rising from 2000 to exceed the atmospheric parameter by 2007. This behaviour is quite closely mirrored in the differences between observed and anticipated interhemispheric trends in Fig. 4 (but starting earlier because of the presumed equatorial location of vegetation that is influenced by Mt. Pinatubo eruption, thus the interhemispheric gradient is not significantly perturbed).

After 2007, slowing emissions lead to a slowing in \(\frac{dC_e}{dt}\), e.g. in Fig. 6b, such that the good agreement before 1990 (see Fig. 2) is again achieved, and the interhemispheric gradient is consistent with this. It is possible that this includes a CO\(_2\) response to the 2007–2009 Global Financial Crisis (Le Quéré et al., 2009). However the situation will be better evaluated with improved time resolution and sensitivity to growth rate calculation away from end-effects, and when the behaviour of the current ENSO cycle is better defined.

6.1. Fluxes required to reconcile atmosphere and emission trends

To estimate the magnitude of fluxes (anthropogenic or natural) required resolving the apparent trend conflicts over the last two decades, we can quantify differences between \(\frac{dC_e}{dt}\) and \(\frac{dC_a}{dt}-EV\) as discrepancies in the source estimates or departures from modelled sink behaviour. Figure 7 reproduces results from Fig. 6, but presenting annual average values in PgC yr\(^{-1}\), taken from 1994.5 to avoid the early-1990s volcanic influence. Figure 7b shows the emissions required to match \(\frac{dC_a}{dt}-EV\) in Fig. 7a, assuming ongoing \(C_e\)-dependent ocean and terrestrial modelled uptake (each steadily increasing at \(\sim 0.04\) PgC yr\(^{-1}\) yr\(^{-1}\)). For convenience we present adjustments to emissions, but these could also represent the magnitude of global
DIFFERENCES BETWEEN ATMOSPHERIC CO₂ AND ANTHROPOGENIC CO₂ EMISSIONS

Fig. 7. (a) Shows annual values of measured atmospheric growth adjusted for EV in PgC yr⁻¹ obtained from the cga monthly data in Fig. 6a. The modelled dC/dt using reported emissions, and the long-term mean SPO trend are included for comparison. (b) Uses the model to calculate emissions PgC yr⁻¹ (filled symbols) that are consistent with the atmospheric measurements from cga; emissions calculated using mlu (open symbols) are also included for comparison. The –1 PgC yr⁻¹ adjustment to the cga curve addresses high northern latitude emissions in 2002–2003 that are not captured by the predominantly equatorially forced EV index (see text). These exceed the reported emissions (open squares) between 1994 and 2004.

sink anomalies that are required for consistency with reported emissions. The 2002/2003 peak is not well captured in the predominantly equatorially forced empirical EV. The adjusted line with open symbols in Fig. 7b illustrates the result of attributing extra emissions of 1 PgC yr⁻¹, spread over 2002 and 2003 for the dC/dt-EV₂ case, reducing the difference from dC/ dt in this period. After 2003, and in the absence of independently documented sink anomalies, dC/ dt decreases on average from the peak while reported emissions continue to increase strongly until 2007, before declining to agree with the four-decade SPO trend by 2009.

Between 1994 and 2003, the difference between the reported and modelled emissions (for the adjusted dC/dt-EV₂) is 5.1 PgC over 10 yr, and, for example, is the largest sustained difference between the dC/dt and the MLO, SPO linear trend lines in Fig. 2. If interpreted as an error in emissions, the difference over the 1994–2003 period would represent an underestimation of 7% in the fossil fuel and cement emissions (reported total 67 PgC), or 34% of the reported LUC component (15 PgC), that is about 6% in total anthropogenic emissions. Alternatively, the combined modelled ocean and terrestrial sinks used in estimating dC/dt (49 PgC) are 20% overestimated.

Trends in global sink behaviour are often discussed in terms of changes in airborne fraction (the proportion of total anthropogenic emissions that remain in the atmosphere, e.g. Raupach et al., 2008). In determining trends and uncertainties in airborne fraction behaviour, it is important to acknowledge the possible sampling and emission biases discussed in this paper. Assuming that the measures adopted above effectively remove bias, the model airborne fraction using adjusted annual emissions to fit the dC/dt-EV₂ is near-constant after 1965 at 0.42 ± 0.02% (and for example, is 0.41 ± 0.02% after 1990).

6.2. Decadal sink changes

If the relatively stable estimates of emissions through the 1990s is correct, and is due to slowing of NH emissions, then our suggested combined atmospheric constraints of global growth trends and interhemispheric gradient trend require a northern non-fossil source of carbon (~20% above the historic values given by the Trudinger et al. (1999) model for the period) to maintain the interhemispheric gradient.

For the period of acceleration in the post-2000 reported emissions, a rapidly increasing northern sink might explain the absence of change in the interhemispheric gradient, but would not provide sufficient net CO₂ to explain the recent observed annual growth rates. An effective source (reduced sink) in the Southern Hemisphere (SH) that balances interhemispheric gradient, would lead to recent atmospheric growth rates significantly above those observed, unless there is a coordinated equatorial sink. In that context, the absence of reported volcanic eruptions with VEI ≥ 5 since 1991 and low ENSO activity after 2000 facilitates this analysis.

Le Quéré et al. (2007) use ocean general circulation modelling plus inversion of southern atmospheric CO₂ data and find a slowing Southern Ocean CO₂ uptake of around 0.08 PgC yr⁻¹ per decade, and this is supported in sign and magnitude by changes in South Indian Ocean fugacity measurements relative to the atmospheric change (Metzl, 2009). While the modelling results have been challenged (e.g. Boning et al., 2008; Law et al., 2008) and the fugacity measurements have large uncertainty and uneven temporal structure, even if correct, the magnitude of the flux changes is of marginal influence in the global context under discussion here. Also, the suggested sink decrease in the north Atlantic gyre (Watson et al., 2009) is in the wrong sense to help resolve the interhemispheric problem highlighted here. In terms
of magnitude, the post-2000 atmospheric growth attributable to reported emissions increases at 2.1 PgC yr\(^{-1}\) per decade, while our modelled global sinks increase at 0.9 PgC yr\(^{-1}\) per decade (approximately equally distributed between land and oceans).

More recently, Sarmiento et al. (2009) report an increase in the global terrestrial biosphere sink by 0.88 PgC yr\(^{-1}\) (~0.4 PgC yr\(^{-1}\) per decade) after 1988/1989 and decrease in global ocean sink by 0.35 PgC yr\(^{-1}\) (~0.2 PgC yr\(^{-1}\) per decade), reducing atmospheric accumulation by 0.53 PgC yr\(^{-1}\) over the same period (~0.3 PgC yr\(^{-1}\) per decade). Their conclusion about large increases in total land sink is based on similar emission estimates and NH atmospheric data (i.e. susceptible to misinterpretation of regional fluxes as being global) to those used by Le Quéré et al. (2009), but with partitioning between source/sink processes determined by the model based estimate of ocean sink. Bottom up estimates of land and ocean fluxes remain too uncertain to contribute to reliable estimates of global trends (e.g. their bottom-up estimate of total land sink change from 1988 to 1989 is equivalent to 1.1 ± 1.2 PgC yr\(^{-1}\)). Our measured atmospheric growth, adjusted for ENSO and volcanic influence, provides higher time-resolution than that described by Sarmiento et al., increasing at the historic rate of ~0.5 PgC yr\(^{-1}\) per decade through the 1990s and slowing by ~0.02 PgC yr\(^{-1}\) per decade after 2002/2003.

An accelerating terrestrial sink of the magnitude suggested by Sarmiento et al. (2009), constrained to be at mid-to-high northern latitudes by our suggested interhemispheric gradient constraint, raises serious concerns about the ability of the terrestrial sink to maintain such high rates into the future. However, there are other possible explanations of the atmospheric data.

6.3. Anthropogenic emission estimates

The Sarmiento et al. result is based on a 5% uncertainty in fossil fuel emissions which, while it may represent the present situation, significantly underestimates uncertainty in the 1990s (G. Marland and R. Andres, personal communication 2009). Recently, van Vuuren and Riahi (2008) have examined the quality of anthropogenic emissions inventory data, suggesting ‘the recently observed increase in the rate of emission growth could very well turn out to be a statistical artefact’, based primarily on uncertainties in Chinese emissions. Our suggested underestimation of 1990s emissions in order to reconcile modelled and observed growth from the 1990s is well within a 10–15% uncertainty of early emission estimates and offers a possible simple explanation for the surge in reported emissions, being a consequence of connecting systematic early underestimates (more subject to possible economic and/or political bias? or evolving methodology), with more accurate recent estimates.

7. Conclusions

The trend in global atmospheric CO\(_2\) growth, thus in CO\(_2\)-induced global warming rate, is most reliably indicated in the large homogeneous CO\(_2\) concentration fields in the troposphere south of 30 S. Annual accumulation of CO\(_2\) in the atmosphere has marginally slowed after climate-induced high-northern-latitude peak emissions in 2002/2003, to fall below the four-decade baseline defined by a linear regression through Mauna Loa and South Pole growth-rate data (but still around ~2 ppm yr\(^{-1}\) in 2009). Even if the global nature of our trends is contested, the ability of models to represent the precisely determined behaviour of such a large portion of the global troposphere is a strong and not-yet-adequately applied constraint on global budgeting models. A pressing requirement for abatement of anthropogenic climate forcing due to CO\(_2\) is that the dC\(_2\)/dr approaches zero.

Adjustment for the historic (mainly equatorial) ENSO and volcanic influences (EV) on dC\(_2\)/dr suppresses large natural perturbations during the 1990s, achieving consistency with the four-decade linear growth trend. The EV correction is small and generally well correlated with CO\(_2\) growth after 2002/2003, such that it does not contribute significantly to the 2003–2009 slowing in growth. A possible further slowing from 2008 may be indicative of reduced anthropogenic emissions associated with the Global Financial Crisis (Le Quéré et al., 2009), though uncertainty near the end of the smoothed records cannot yet exclude an ENSO-induced influence (but future better definition of the current ENSO cycle may do this).

The decadal behaviour contrasts with modelled atmospheric trends, dC\(_2\)/dr, attributable to fossil and land-use emissions using historically consistent C\(_2\)-dependent parametrizations of oceanic and terrestrial sinks. The dC\(_2\)/dr flatten through the 1990s and accelerate rapidly after 2000. Consistency with past dC\(_2\)/dr growth (due mainly to NH emissions) suggests there should be proportional increase in the interhemispheric CO\(_2\) difference, which is not observed. In the absence of firm evidence for sufficiently large systematic bias in 1994–2003 terrestrial and/or oceanic sinks, systematic underestimation of emissions of around 6% is a simple and likely explanation for the dC\(_2\)/dr and dC\(_2\)/dr-EV trend differences. In this scenario, the recent responses of combined terrestrial and oceanic sinks to increasing atmospheric CO\(_2\) remain broadly consistent with historic behaviour, and provide no evidence for volatility in the carbon cycle on a global scale. Alternatively, complex and unusual sink behaviour is required.

An approach that has been suggested for studying the recent dC\(_2\)/dr trend, is to seek statistical precedents for the observed trends. Although there have been periods in the past where dC\(_2\)/dr has decreased, a purely statistical approach such as this does not take into account available information on the causes for variations in the dC\(_2\)/dr. For example, dC\(_2\)/dr decreased around the early 1990s, but Pinatubo provides a likely explanation. The recent dC\(_2\)/dr decrease coincides with an unprecedented increase in reported anthropogenic forcing, and to date we have no independent information on natural sinks that would explain the observed decrease. Increasing global vigilance for anomalous
REGIONAL CARBON FLUXES LIMITS POSSIBILITIES FOR UNDETECTED NATURAL CONTRIBUTIONS. UNDER THESE CIRCUMSTANCES, MEASUREMENTS FROM CONTINUOUS MONITORING OF SELECTED MID-TO-HIGH LATITUDE SH MARINE BOUNDARY-LAYER AIR OFFERS READILY ACCESSIBLE ON-GOING STRONG CONSTRAINTS ON GLOBAL CO2 EMISSIONS.

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Supporting Information

Additional supporting information may be found in the online version of this article.

Appendix S1: Selection of marine boundary layer air at Cape Grim.

Appendix S2: Growth rate error from measurement bias.

Appendix S3: ENSO and volcanic influence on CO2 growth rate.

Appendix S4: Modelled response to emission estimates.

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