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**Airborne fraction  
trends and carbon  
sink efficiency**

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# What can be learned about carbon cycle climate feedbacks from CO<sub>2</sub> airborne fraction?

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## Abstract

The ratio of CO<sub>2</sub> accumulating in the atmosphere to the CO<sub>2</sub> flux into the atmosphere due to human activity, the airborne fraction (AF), is central to predict changes in earth's surface temperature due to greenhouse gas induced warming. This ratio has remained remarkably constant in the past five decades, but recent studies have reported an apparent increasing trend and interpreted it as an indication for a decrease in the efficiency of the combined sinks by the ocean and terrestrial biosphere. We investigate here whether this interpretation is correct by analyzing the processes that control long-term trends and decadal-scale variations in AF. To this end, we use simplified linear models for describing the time evolution of an atmospheric CO<sub>2</sub> perturbation. We find firstly that the spin-up time of the system for the AF to converge to a constant value is on the order of 200–300 years and differs depending on whether exponentially increasing fossil fuel emissions only or the sum of fossil fuel and land use emissions are used. We find secondly that the primary control on the decadal time-scale variations of the AF is variations in the relative growth rate of the total anthropogenic CO<sub>2</sub> emissions. Changes in sink efficiencies tend to leave a smaller imprint. Before interpreting trends in the AF as indication of weakening carbon sink efficiency, it is therefore necessary to account for these trends and variations, which can be achieved based on a predictive equation for the AF implied by the simple models. Using atmospheric CO<sub>2</sub> data and emission estimates for the period 1959 through 2006 we find that those controls on the AF, omissions in land use emissions and extrinsic forcing events can explain the observed trend, so that claims for a decreasing trend in the carbon sink efficiency over the last few decades are unsupported by atmospheric CO<sub>2</sub> data and anthropogenic emissions estimates.

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# 1 Introduction

Central for predicting future temperatures of the Earth's surface is how much and for how long carbon dioxide from fossil fuel emissions and land use change stays in the atmosphere, and how much gets removed by the carbon sinks on land and in the ocean (e.g., Solomon et al., 2009). A straightforward measure of this redistribution is the ratio between the increase rate in atmospheric CO<sub>2</sub> and the CO<sub>2</sub> emitted to the atmosphere by human activity (fossil fuel burning and land use change). Keeling (1973) termed this quantity the "airborne fraction" (AF) and it was investigated in many subsequent studies (e.g. Bacastow and Keeling, 1979; Oeschger and Heimann, 1983; Enting, 1986). Because of the large uncertainties in land fluxes, these early studies could estimate the value of AF only to within a wide range from 0.38 to 0.78 (Oeschger and Heimann, 1983). Recently several studies have extended the estimation of AF over the last two decades, with a suggestion of a positive trend in AF (Canadell et al., 2007; Raupach et al., 2008; LeQuere et al., 2009). Moreover, this positive trend has been interpreted as evidence for a decreasing trend in the efficiency of the ocean and land carbon sinks. Given the model-based projection of a substantial reduction in the sink strength of the ocean and land in the future (e.g. by a large-scale dieback of the Amazon old-growth forest, Cox et al., 2000), the notion that the sinks have already begun to deviate from a linear response to the atmospheric CO<sub>2</sub> perturbation is a source of substantial concern. While there remains discussion about whether this trend in the AF is actually statistically significant (Knorr, 2009), we focus our discussion here on whether the inferred conclusion is possible, i.e. whether an increasing trend in the AF implies a decreasing efficiency of the carbon sinks.

Determinants of the AF are the magnitude and time course of the human induced emissions of CO<sub>2</sub> into the atmosphere and the removal of this anthropogenic carbon by the ocean and land biosphere. It has been known since the early 1970's, possibly earlier, that the AF will eventually asymptote to a constant value if (i) the CO<sub>2</sub> uptake by the oceans and land ecosystems is linear and (ii) if CO<sub>2</sub> emissions to the

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atmosphere follow exactly an exponential function (e.g., Bacastow and Keeling, 1979). Thus, given that fossil fuel emissions have risen approximately exponentially over the last 250 years, and that natural systems tend to respond linearly to small perturbations, it is natural to inquire whether time trends in AF may inform us about changes in the linear behavior of carbon uptake by the oceans and land ecosystems (Canadell et al., 2007; Raupach et al., 2008; LeQuere et al., 2009; Rafelski et al., 2009). However, the relative growth rate  $RGR \equiv \frac{1}{FF} \frac{dFF}{dt}$  of fossil fuel emissions FF has varied by more than a factor of two in the last 100 years (e.g., Raupach et al., 2008). In addition, emissions from land use change exhibited an even more varied time course, so that the total emissions only very approximately followed a single exponential. Furthermore trends may be an articulation of incomplete spin-up of the system for the AF to reach a stationary value. We examine here the impact of these deviations and controls on the AF, and what the consequences are for the interpretation of the AF as an indicator for changes in the efficiency of carbon sinks, and in turn the state of the global carbon cycle.

Before proceeding, it is important to recognize that definitions of the AF in the literature vary. Studies from the 1970s and 1980s defined airborne fraction from cumulative carbon inventory changes as

$$\frac{C(t_f) - C(t_i)}{\int_{t_i}^{t_f} FF(t) dt}$$

(Keeling, 1973; Bacastow and Keeling, 1979; Enting, 1986) or alternatively as

$$\frac{C(t_f) - C(t_i)}{\int_{t_i}^{t_f} FF(t) + LU(t) dt}$$

(Oeschger and Heimann, 1983) where  $t_i, t_f$  are the beginning and the end time of the period considered, FF is fossil fuel emissions and LU is the flux to the atmosphere due to land use change. The more recent studies define airborne fraction from annual or

monthly inventory changes as either

$$AF_{FF} \equiv \frac{\frac{dC}{dt}(t)}{FF(t)}, \text{ or } AF_{FF+LU} \equiv \frac{\frac{dC}{dt}(t)}{FF(t) + LU(t)}$$

(Canadell et al., 2007; Raupach et al., 2008; LeQuere et al., 2009; Knorr, 2009). We analyze here the time-evolution of the AF as defined by these recent studies.

As our analysis is intricate, reflecting the complicated nature of the problem, we briefly outline the organization of our paper. We start in Sect. 2 with a characterization of the time course of anthropogenic CO<sub>2</sub> emissions and carbon sinks, thereby highlighting that there have been strong variations in the relative growth rate of fossil fuel emissions over the last century. In Sect. 3 we introduce a simple linear model of the evolution of an atmospheric CO<sub>2</sub> perturbation, thereby also clarifying the meaning of “sink efficiency”. In Sect. 4 we explore how the time course of the anthropogenic emissions controls variations in the AF, using a predictive equation implied by our simple model. We demonstrate that (i) for an atmospheric CO<sub>2</sub> perturbation which is not following an exact exponential function, there is an adjustment time for the AF to converge to its constant asymptotic value which is on the order of centuries, and that (ii) variations in the relative growth rate of the anthropogenic emissions are a major control on variations of the AF. Therefore, in order to unravel trends in the AF caused by trends in carbon sink efficiency or extrinsic non-anthropogenic events, like volcanic eruptions, signatures due to incomplete “spin-up” and fossil fuel growth rate variations need first to be removed from the observed AF. We can achieve this using our predictive equation for the AF. We then examine the remaining signal for trends not explained by known extrinsic non-anthropogenic forcings or omissions in anthropogenic fluxes, to conclude whether there is indeed evidence for carbon sink efficiency trends in the observed AF record. This terminates our main analysis. Section 5 in addition explores the signal to noise ratio of AF trends caused by sink efficiency trends, and finally we conclude.

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## 2 Anthropogenic carbon sources and sinks

The main driver of the rapid increase in atmospheric CO<sub>2</sub> is fossil fuel emissions, which are estimated from national energy statistics with an uncertainty of ~5% (Marland, 2006, updated by Boden et al., 2009). A logarithmic representation (Fig. 1c) reveals that fossil fuel emissions have increased roughly exponentially, with the time-scale of relative change, the inverse of the relative growth rate, varying roughly between ~20 and 150 years (Fig. 1e). The time-scale  $\tau_f$  of relative change of an anthropogenic flux  $f$  to the atmosphere is defined as its logarithmic derivative:

$$\tau_f \equiv \left(\frac{1}{f} \frac{df}{dt}\right)^{-1} \simeq \left(\frac{1}{f} \frac{\Delta f}{\Delta t}\right)^{-1}$$

with  $\Delta f \equiv f(\text{yr} + 1) - f(\text{yr})$  and  $\Delta t = 1$  year. Variations in the time-scale of relative change of fossil fuel emissions are mainly due to economic cycles and wars. Thus there was an approximately 80 year period from around 1830 to 1910 (approximately the start date of World War one (WW1)) with a roughly constant  $\tau_{\text{FF}} \equiv \left(\frac{1}{\text{FF}} \frac{d\text{FF}}{dt}\right)^{-1} \simeq \left(\frac{1}{\text{FF}} \frac{\Delta\text{FF}}{\Delta t}\right)^{-1}$  of 20 years (equivalent to a relative growth rate of 5% per year, Fig. 1e). The WW1, post WW1 and great depression period saw less growth, with both positive and negative time-scales resulting in a substantially longer mean  $\tau_{\text{FF}}$ . After WW2 (starting around 1948) there is again fast growth, paralleling the recovery of industrial countries' economies, until the early 1970s with  $\tau_{\text{FF}}$  of ~20 years. From the early 1970s until ~1999 the  $\tau_{\text{FF}}$  increased again to ~80 years (relative growth rate of ~1.3% yr<sup>-1</sup>). Growth has returned close to the 1830 to 1910 and post WW2 values starting around 2001.

The second cause of the rise in atmospheric carbon, and at the same time the least well constrained component of the atmospheric carbon budget, is carbon fluxes released from land to the atmosphere due to land use change (for example rainforest to pasture conversion in the tropics, or peat burning 1997/1998 in Indonesia as a consequence of attempts to convert swamp forests to rice paddies at a large spatial scale, Page et al., 2002). Estimates of Houghton et al. (2007) indicate that this term has also

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risen in time, but at a considerably smaller rate than fossil fuel emissions (Fig. 1d). The uncertainty in fluxes associated with land use change is large, on the order of 40–100%, as revealed by the range of published estimates (e.g. Grainger, 2008; Houghton et al., 2007; DeFries et al., 2002; Achard et al., 2002).

The atmospheric CO<sub>2</sub> accumulation rate is well constrained by atmospheric concentration records (Keeling, 1960; Etheridge et al., 1996). Estimates of ocean uptake of anthropogenic carbon based on various methods have also converged over recent years to 2.2±0.2 PgC yr<sup>-1</sup> for a nominal period of ~1995–2000 (Sabine et al., 2004; Sweeney et al., 2007; Sarmiento et al., 2009; Gruber et al., 2009; Khatiwala et al., 2009). The net land sink, the sum of the land sink and the CO<sub>2</sub> flux to the atmosphere due to land use change, can then be calculated as the difference between fossil fuel emissions, the atmospheric CO<sub>2</sub> accumulation rate and ocean uptake. The implied net land sink stayed roughly constant with a mean value of nearly zero from the 1930s to 1990 and then increased to a magnitude of approximately 1 PgC yr<sup>-1</sup> for the 1990s and early 2000s (e.g., Sarmiento et al., 2009).

### 3 A simple carbon cycle model

We now introduce how we attempt to analyze the problem. First we need to formalize the concept of a “sink efficiency”. An efficiency, like the efficiency of a heat engine, is defined as the ratio between the magnitude of an effect and the magnitude of its cause. Thus the concept of sink efficiency is tied to a linear description of the effect-cause relationship. In our case the cause is the increase in atmospheric CO<sub>2</sub> due to human activity and the effect is the carbon flux from the atmosphere to the oceans and land carbon pools. Thus, for constant sink efficiency, the fluxes from the atmosphere to the oceans and to the land,  $F_{at \rightarrow oc}$  and  $F_{at \rightarrow ld}$ , are given by

$$F_{at \rightarrow oc} = \frac{\Delta C}{\tau_{oc}}, F_{at \rightarrow ld} = \frac{\Delta C}{\tau_{ld}}, \quad (1)$$

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where  $C$  is atmospheric carbon dioxide,  $\Delta C \equiv C(t) - C(1765)$  is the anthropogenic perturbation of atmospheric carbon dioxide, and  $\tau_{oc}$  and  $\tau_{ld}$  are strict constants. In this context, a weakening/strengthening of the sinks then means that  $\tau_{oc}$  and/or,  $\tau_{ld}$  are increasing/decreasing in time.

Given that the concept of “sink efficiency” is tied to a linear description, the approach we take here is to investigate what such a linear model of the evolution of an atmospheric  $\text{CO}_2$  perturbation will tell us about the controls on variations and trends in airborne fraction. The simplest linear model based on the mass balance of atmospheric carbon  $C$  is given by

$$\begin{aligned} \frac{d\Delta C}{dt} &= f(t) - F_{at \rightarrow oc} - F_{at \rightarrow ld} = \\ &= f(t) - \left( \frac{1}{\tau_{oc}} + \frac{1}{\tau_{ld}} \right) \Delta C = f(t) - \frac{\Delta C}{\tau_s}. \end{aligned} \quad (2)$$

Here  $t$  is time, the subscript  $s$  stands for “system”,

$$\frac{1}{\tau_s} \equiv \frac{1}{\tau_{oc}} + \frac{1}{\tau_{ld}}$$

is the proportionality constant between the atmospheric  $\text{CO}_2$  perturbation and the total C flux out of the atmosphere, and  $f(t)$  is the anthropogenic  $\text{CO}_2$  flux into the atmosphere, which we can view as forcing of the system. For our problem  $f$  is mostly FF + LU although we will also consider the case of  $f = \text{FF}$  alone.

It is interesting to consider to what extent the assumption of a linear relationship between the flux out of the atmosphere and the anthropogenic atmospheric  $\text{CO}_2$  perturbation is justified. In the case of the ocean uptake flux, this assumption is actually rather well justified, because the driving force for the uptake is the air-sea  $\text{CO}_2$  disequilibrium (e.g. Sarmiento et al., 1992; Maier-Reimer and Hasselmann, 1987). This is also confirmed by 3-D ocean model simulations (e.g., Mikaloff-Fletcher et al., 2006), although such simulations also show a strong deviation from linearity once atmospheric  $\text{CO}_2$



has risen to values where the surface ocean buffer factor begins to change strongly (Sarmiento and LeQuere, 1996). Specifically using this modelling result and the anthropogenic ocean carbon inventory estimated from oceans surveys for 1995 (e.g., Gruber et al., 2009), we obtain a rough estimate of  $\tau_{OC} \simeq 81.4$  yr (Appendix C).

For uptake by land vegetation it is less clear whether the linearity concept applies. This is because uptake by land, unlike the oceans, is tied to productivity and the status of the land vegetation. Some of these processes may possibly be related to the atmospheric  $\text{CO}_2$  perturbation (specifically  $\text{CO}_2$  uptake during photosynthesis), while others like nutrient and micronutrient availability, plant and soil respiration, vegetation population dynamics, and land use change are not. Even if there were a productivity increase due to  $\text{CO}_2$  “fertilization”, it would likely be a linear response only during a limited period of time until land vegetation reaches a new steady state balance between growth and mortality. The linear response assumption of the land vegetation thus confounds many processes and time-scales (e.g., Lloyd, 1999).

One may question the realism of our simple model on the grounds that the model treats both the oceans and the land vegetation as just one integral pool, while at least for the oceans several pools with characteristic exchange time-scales seems more realistic. However it turns out that inclusion of multiple ocean pools does not alter our conclusions (see discussion below and Appendix D).

#### 4 Predicted and observed evolution of airborne fraction

To get a general sense of the implications of this model (Eq. 2) for the time-course of AF, we have calculated AF for three idealized forcing functions  $f(t)$ : (i) exponential forcing  $f(t) = f e^{t/\tau_f}$  with a single characteristic time-scale  $\tau_f$  (or equivalently relative growth rate  $1/\tau_f$ ); the subscript  $f$  refers to “forcing”, (ii) the sum of an exponential function and a constant, and (iii) the sum of several exponential functions with different characteristic time-scales or relative growth rates (Appendix A and Fig. 2). The first case is an idealization of forcing the atmosphere with fossil fuel burning  $\text{CO}_2$  alone,

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while the latter two cases mimic forcing of the atmosphere with the sum of fossil fuel emissions and land use change emissions.

As shown previously (e.g., Bacastow and Keeling, 1979), AF is constant for a purely exponential forcing (Appendix A and Fig. 2). For forcing functions, which differ from an exact exponential function, AF converges to an asymptotic value after some spin up time. For all forcings the asymptotic value of AF is given by

$$AF = \frac{1}{1 + \frac{\tau_f}{\tau_s}}$$

and is thus controlled by the ratio between the forcing time-scale and the system response time-scale. For the case of forcing by the sum of several exponentials it is the  $\tau_f$  of the fastest growing exponential function that determines the asymptotic value of AF (see last equation in Appendix A). If the forcing is not exactly exponential, then the time-scale for convergence is roughly on the order of 200–300 years (Fig. 2), depending a bit on the exact functional form of the forcing.

An intuitive explanation for the existence of a spin-up period is as follows. The constancy of AF for a purely exponential forcing reflects the balance between two exponential processes, exponential damping of the atmospheric perturbation via carbon sinks and exponential forcing (Appendix B). If the forcing deviates from a pure exponential function, there will be a spin up period until the exponential component of the forcing dominates over other slower growing components of the forcing. An implication of the existence of a spin-up period is that we expect observed  $AF_{FF+LU}$  to converge towards  $AF_{FF}$  from lower values. This is because fossil fuel emissions rise approximately exponentially but land use change emissions rise more slowly and thus their sum will not equal an exact exponential function (Sect. 2).

Instead of idealized cases we now predict the time-course of AF using the observed FF and LU emissions. For this purpose we use the differential equation for AF implied by our model Eq. (2)

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$$\frac{dAF}{dt} = \left( \frac{1}{f} \frac{df}{dt} + \frac{1}{\tau_s} \frac{d\tau_s}{dt} \right) - \left( \frac{1}{\tau_s} + \frac{1}{f} \frac{df}{dt} + \frac{1}{\tau_s} \frac{d\tau_s}{dt} \right) AF \quad (3)$$

derived in Appendix B. This equation shows that the relative growth rate of the forcing, or equivalently the inverse of the time-scale of relative change of the forcing,

$$\frac{1}{\tau_f(t)} \equiv \left( \frac{1}{f(t)} \frac{df(t)}{dt} \right),$$

5 and the relative change of the time scale of the combined land and ocean response,

$$\frac{1}{\tau_s} \frac{d\tau_s}{dt},$$

control trends in AF.

To predict the variations in AF according to our simple model, we integrate the equation numerically assuming a constant sink efficiency, i.e.  $\tau_s = \text{const}$ . We choose the value for  $\tau_s$  such that the mean observed and predicted AF are equal over the period 1959-2007 using least squares, which results in  $\tau_s = 42$  years for  $AF_{FF}$  and  $\tau_s = 37.5$  years for  $AF_{FF+LU}$ . The results based on the fossil fuel time-series estimated by Marland (2006) alone, as well as the sum of the fossil fuel and land use time-series, used as forcing, are shown in Figs. 1a and 3a. The AF records calculated from atmospheric concentration data (from Mauna Loa) and anthropogenic emissions are also shown.

To estimate observed  $AF_{FF}$  and  $AF_{FF+LU}$  respectively, we have calculated  $dC/dt$  using the monthly mean records from NOAA ESRL (co2\_mm\_mlo.2009.txt obtained in November 2009 from ftp://ftp.cmdl.noaa.gov/ccg/co2/trends/ Tans and Conway, 2009). From these monthly data we first calculated annual means centered on 31 December/1 January, from which we estimated the time derivative by differencing. We estimate the AF from annual means, because the anthropogenic emissions estimates are annually resolved and for reproducibility of the calculations. Our conclusions are not sensitive to this choice. Besides using the requirement for agreement of the mean AF over the period from 1959 to 2007 to estimate  $\tau_s$ , we may also determine  $\tau_s$  from the

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mass conservation requirement that predicted and observed increase in atmospheric  $\text{CO}_2$  agree. The two estimates agree well (i.e.  $\tau_s \sim 37.5$  years).

Observed  $\text{AF}_{\text{FF}}$  and  $\text{AF}_{\text{FF+LU}}$  records (Figs. 1a, 3a) exhibit large inter-annual variability, which is missing in the  $\text{AF}_{\text{FF}}$  and  $\text{AF}_{\text{FF+LU}}$  predicted by the linear model. This is because our model is only forced by carbon fluxes from fossil fuel and land use change, thus variations due to other indirect, non-anthropogenic forcings, like volcanic eruptions or climate oscillations, are not captured. The large inter-annual variations in observed air-borne fraction are largely due to inter-annual variability in the rate of change of atmospheric  $\text{CO}_2$ ,  $\frac{dC}{dt}$ , an observation known since the seventies to be associated with El Niño/La Niña and post volcanic periods (Agung, El Chichon, Pinatubo; Bacastow, 1976).

The forcing during the period 1959–2007 has three distinctly different phases (1958–1973 fast growth, small  $\tau_f \sim 20$  yr; 1973–1999 slow growth, large  $\tau_f \sim 30$ –150 years; 2000–2006 fast growth, small  $\tau_f \sim 25$  years; Fig. 1b, e). We thus expect predicted and observed AF to be lower during the 1973–1999 period compared to the other two periods, with transition periods in between (irrespective of considering  $\text{AF}_{\text{FF}}$  or  $\text{AF}_{\text{FF+LU}}$ ). This is indeed what we find (Figs. 1a, 3a). Generally for this model it is also evident from Fig. 1 a and b that there is indeed a tight relation between AF and relative growth rate  $\text{RGR} = \frac{1}{\tau_f}$  of anthropogenic emissions. As mentioned earlier on, because the forcing used to calculate  $\text{AF}_{\text{FF+LU}}$  is approximately the sum of an exponential function (FF) and a less strongly increasing function (LU), we expect  $\text{AF}_{\text{FF+LU}}$  (red curve) to be lower than  $\text{AF}_{\text{FF}}$  (blue curve) and to slope more upwards than  $\text{AF}_{\text{FF}}$ , eventually converging towards  $\text{AF}_{\text{FF}}$ . This is indeed what is observed and predicted (Fig. 1a).

Given the variation in  $\text{AF}_{\text{FF}}$  and  $\text{AF}_{\text{FF+LU}}$  due to variations in forcing (Figs. 1a, 3a), particularly in fossil fuel emissions, and the considerable time it takes for AF to converge to its asymptotic value, is there nonetheless a possibility to test whether there are trends in sink efficiency from the time course in AF? If our differential equation for AF based on the assumption of a linear response were to fit the data well, then we would not need to invoke a trend in sink efficiency (i.e. a trend in  $\tau_s$ ). The difference (resid-

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uals) between observed and predicted AF can thus give us an indication on potential non-linearities or possibly incompleteness of the linear model to describe the evolution of the anthropogenic atmospheric carbon perturbation. The trend of the residuals (the difference between observed and predicted  $AF_{FF+LU}$ ) is positive (Fig. 3d), indicating that something is indeed at odds. There could be three causes for the trend in the residuals: (i) incomplete forcing, particularly the absence of forcings associated with indirect, i.e. non-anthropogenic mechanisms such as volcanic eruptions, (ii) the response time scale (or equivalently sink efficiency) could be changing, or (iii) the model could be all too simplistic.

We may get some insight into what causes the trend in the residuals by inquiring what corrections  $\Delta f$  to FF + LU would be needed to obtain a better fit between observed and predicted AF. If we can attribute these flux corrections to extrinsic non-anthropogenic forcings or omissions in land use and fossil fuel fluxes, then there is no need to invoke trends in sink efficiency and vice versa. To estimate the flux corrections  $\Delta f$ , we minimize

$$J(\Delta f(1959), \dots, \Delta f(2006)) = \sum_{yr=1959}^{2006} (AF_{FF+LU}^{obs}(yr) - AF_{FF+LU}^{pred}(yr))^2 + ((C_{2006}^{pred} - C_{1959}^{pred}) - (C_{2006}^{obs} - C_{1959}^{obs}))^2$$

with respect to  $\Delta f(1959), \dots, \Delta f(2006)$  using simulated annealing. The second term of  $J$  ensures that mass is conserved. Because the weighting of the data is uniform, there should not be a significant trend in the residuals after including the flux corrections. To be sure, we used the standard t-test (e.g. Robinson, 1981 and Appendix E) to test whether there is a significant slope in the residuals after including the flux corrections and found indeed no significant slope.

The estimation procedure identifies four events (Fig. 3c): increased sinks for atmospheric carbon in the aftermaths of the 1963 Agung and 1991 Pinatubo eruptions and

carbon flux pulses to the atmosphere in 1997/1998 and similarly in 2002/2003. A dip in the increase rate of atmospheric carbon is well known to occur after major volcanic eruptions, especially those that inject material into the stratosphere (e.g., Rödenbeck et al., 2003). The decrease in atmospheric CO<sub>2</sub> is generally attributed to a land sink in the aftermath of the eruption. The mechanism may possibly be an increase in the ratio between diffuse and direct radiation, enhancing photosynthesis (Roderick et al., 2001) and/or reduced soil respiration due to temporary cooling of the earth surface (Jones and Cox, 2001). The 1997/1998 carbon flux pulse to the atmosphere is also well studied, and largely attributed to peat burning from Indonesia in 1997/1998 (Page et al., 2002). This carbon flux to the atmosphere seems to be missing from the Houghton et al. (2007) land use change flux estimate, although it is the result of land use change (Page et al., 2002). Finally there are indications from several studies of what the causes of the 2002 and 2003 flux pulses to the atmosphere could be (Yurganov et al., 2005; Balzter et al., 2005; Jones and Cox, 2005). Specifically Yurganov et al. (2005) documented air-column CO anomalies on the order of 50% at northern hemisphere mid-to high latitude stations, with anomalies occurring during the second half of the year 2002 and 2003. They associated these signatures with boreal forest fires in Siberia, consistent with results from remote sensing fire spot data, and results based on more refined remote sensing methods (Balzter et al., 2005). Besides boreal forest fires, the 2002/2003 events may also be related to the drought in Europe in summer 2003, which reduced net primary production of the land vegetation (Ciais et al., 2005), although decreases in primary production are likely to be paralleled by compensating anomalies in respiration. Thus overall, with the possible exception of the 2002/2003 event, the four events in the residuals can be attributed to omissions in land use change fluxes and extrinsic forcings.

We may then test whether there is a declining trend in sink efficiency by applying the same standard t-test as before to the slope of  $\Delta f$  but with the post-Agung, post-Pinatubo, Indonesian peat pulse and 2002/2003 events excluded, as indicated by the blue dashed line in Fig. 3 c. The result of the t-test is negative. Thus when removing

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the four events there is no evidence for a sink efficiency trend in the AF. Our analysis is ambiguous regarding the possible sudden “positive feedback event” in 2002/2003. However, since forest fires are at least partially part of a natural cycle of boreal forest succession (Wirth et al., 2002; Mollicone et al., 2002), measurements of future net carbon fluxes in this region are necessary to determine to what extent these fluxes are anomalous.

In order to examine the effect of including multiple linearly coupled ocean pools instead of just one pool in our analysis, we may generalize our predictive Eq. (3) for the AF. To do so we replace the single ocean uptake Greens function  $G(t, t') = e^{-(t-t')/\tau_{oc}}$  by the sum of several Greens functions representing the response of different pools with characteristic time-scales of  $\tau_0 = \infty$ ,  $\tau_1 = 433.3$  yr,  $\tau_2 = 83.9$  yr,  $\tau_3 = 11.2$  yr,  $\tau_4 = 0.8$  yr (see Appendices B and D). Solving the generalized predictive equation for AF numerically, we find nearly the same results for AF, confirming that our simple model suffices to analyse the controls on the AF. The reason is that ocean carbon uptake during the 1950–2010 period is primarily governed by one Green’s function, the one associated with  $\tau_2 = 83.9$  yr. Not surprisingly but reassuringly the time-scale is very similar with the time-scale  $\tau_{oc} = 81.4$  yr estimated using ocean anthropogenic carbon inventories, and forward and inverse ocean carbon uptake simulation results based on general circulation models mentioned earlier on (see introduction and Appendix C).

Rafelski et al. (2009) recently analysed atmospheric CO<sub>2</sub> data from 1850 to the present (with pre-1959 data estimated from ice core air enclosures), as well as “Constant airborne fraction anomaly”, the difference between the atmospheric CO<sub>2</sub> record and 57% of the cumulated (time-integrated) fossil fuel emissions using simple carbon cycle models. The use of cumulated fluxes to define “Constant airborne fraction anomaly” differs from our analysis, which focuses on the ratio of instantaneous fluxes (see introduction). They find firstly that the time course of the cumulated difference between the land sink implied by the atmospheric CO<sub>2</sub> balance and their land model predictions resembles the global atmospheric land temperature record. They interpret this result as evidence for a weak positive climate land carbon cycle feedback, which

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is different from what we find, although the time period (1850–2008) they considered is longer than the period we analyse. They also state that the magnitude of the “constant airborne fraction anomaly” from roughly 1920 onwards is unexpectedly small, given the large decadal to multi-decadal variations in the fossil fuel growth rate. They interpret this as evidence that temperature driven land-atmosphere fluxes compensate for the variations in fossil fuel growth rate in the “constant airborne fraction anomaly”, supporting thus their finding of a weak positive climate carbon cycle feedback based on land vegetation models. Unfortunately their study does not estimate the magnitude of the expected changes in “constant airborne fraction anomaly” due to multi-decadal changes in the fossil fuel growth rate, thus it is not possible to assess whether the observed variations are indeed smaller than expected. The study also does not disentangle the signal in “constant airborne fraction anomaly” caused by multidecadal temperature variations alone, and thus it is not possible to assess their hypothesis that the fossil fuel growth rate and climate driven signatures indeed approximately cancel. Altogether therefore it is difficult to compare their results with ours at this stage.

## 5 AF Trends and efficiency of sinks

Although our analysis suggests that the observed variations in the AF primarily reflect changes in the relative growth rate of the total anthropogenic CO<sub>2</sub> emissions and incomplete spin-up of the system, it is still interesting to analyze the relation between trends in sink efficiency and trends in AF within the framework of our simple model. For this purpose we investigate the hypothetical case that  $\tau_s(t) = 42 \text{ yr}$  for  $t < 1959$  and  $\tau_s = 42 \text{ yr} + e \times (t - 1959)$  with  $e = 0.5$ , corresponding to a weakening of the “sink efficiency” by ~50% by the year 2009 compared to 1959, which is equivalent to a quite strong feedback. We then integrate Eq. (3) forward in time, starting from 1765 and compare the result with the record for the AF calculated for constant  $\tau_s$ . Such a weakening

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trend since 1959 would induce a difference in the trend of AF of

$$\frac{\delta AF}{\delta t} \sim 0.1(50 \text{ yr})^{-1} = 0.002 \text{ yr}^{-1}.$$

This shows firstly that a fairly strong positive feedback, operating over a period of 50 years, causes a trend which is roughly of similar magnitude as variations caused by relative growth rate variations in fossil fuel emissions over the 1959–2009 period. Secondly, we may assess the detectability of the signal by applying the t-test for the slope of a regression, mentioned earlier on (e.g. Robinson, 1981 and Appendix E). Given the “natural” variation in AF on the order of 0.15 (Fig. 3a) we find that a 50% sink efficiency decrease over a period of 50 years is detectable at the 10% significance level but not at the 5% significance level. Thus, because firstly a signal of similar magnitude has to be removed from the AF record first, and secondly the “noise” due to extrinsic non-anthropogenic forcings is large, the AF is not a very suitable diagnostic for detecting trends in carbon sink efficiency.

## 6 Conclusions

We have investigated the question of what controls trends and decadal scale variations in CO<sub>2</sub> airborne fraction (AF) using simple linear models describing the evolution of an atmospheric perturbation in CO<sub>2</sub>. Our analysis suggests firstly that variations of the relative growth rate of anthropogenic CO<sub>2</sub> emissions are a major control of variations in AF. Secondly it suggests that there is a spin-up time on the order of 200–300 years for AF to converge to its asymptotic value, if the forcing is not exactly an exponential function, as it is the case for the sum of fossil fuel burning and land use change emissions. A first consequence is that there is no one-to-one association between positive trends in AF<sub>FF+LU</sub> and negative trends in sink efficiency. A second consequence is that in order to detect trends in sink efficiencies from the time course of AF<sub>FF+LU</sub>, it is necessary to disentangle the spin-up time and fossil fuel growth rate variation signatures in the AF from signatures due to other causes. Our differential equation for AF

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permits us to do so by predicting the time course of AF due solely to these two factors. The remaining trends and variations in the residuals are then either due to variations in extrinsic forcings like volcanic eruptions and climate oscillations, omissions in the anthropogenic fluxes to the atmosphere, trends in sink efficiencies, or inadequacies in our model. We do indeed find a positive trend in the residuals. We may investigate whether the trend may be explained by the first two causes just mentioned, by solving for corrections  $\Delta f$  to the anthropogenic fluxes of Marland (2006) and Houghton et al. (2007) such that the predicted AF matches the observed AF and then analysing them. If we may associate extrinsic forcings or omissions in land use change fluxes with the estimated flux corrections, then we do not need to invoke a trend in carbon sink efficiencies to explain the trend in the residuals. The estimated flux corrections  $\Delta f$  reveal four events. From these four events we may associate two with extrinsic non-anthropogenic forcings (volcanic eruptions) and one with an omission in land use change fluxes (peat burning in 1997/1998) in Indonesia. There is only one anomalous growth event in 2002/2003, which we cannot unambiguously attribute to extrinsic forcing or omissions in land use change fluxes. Thus, overall, once we account for known extrinsic forcing events and known omissions in land use change fluxes, we do not need to invoke a long-term trend in carbon sink efficiencies to reproduce the time-evolution of the observed AF. The 2002/2003 event may possibly be a sign of a sudden “positive carbon cycle climate feedback” but at this stage we don’t really know. Finally, although our study provides a proper framework to analyse trends in the AF given controls other than sink efficiency changes, our analysis suggests that trends in AF are not a very good diagnostic to detect changes in carbon sink efficiency because variations in the signal are complex and the signal-to-noise ratio is fairly small.

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## Appendix A

### Solutions of the differential Eq. (2) for idealized cases

In order to calculate the AF for idealized cases we integrate the differential equation

$$5 \quad \frac{d\Delta C}{dt} = -\frac{\Delta C}{\tau_s} + f(t)$$

with initial condition  $\Delta C(-\infty) = 0$  (since  $\Delta C$  is the perturbation of atmospheric carbon).

For purely exponential forcing  $f(t) = f e^{\frac{t}{\tau_f}}$ ,  $\tau_f$  constant, we find by the method of “variation of constant”

$$\Delta C(t) = \frac{f}{\frac{1}{\tau_s} + \frac{1}{\tau_f}} e^{\frac{t}{\tau_f}}$$

10 and thus

$$\text{AF} \equiv \frac{\frac{dC}{dt}}{f e^{\frac{t}{\tau_f}}} = \frac{1}{1 + \frac{\tau_f}{\tau_s}} = \text{constant}.$$

For a forcing of the form  $f e^{t/\tau_f} + f_0$  where  $f_0$  is constant, we may integrate the equation similarly to obtain

$$\text{AF} = \frac{1}{1 + \frac{\tau_f}{\tau_s}} \times \frac{e^{\frac{t}{\tau_f}}}{e^{\frac{t}{\tau_f}} + (f_0/f)}.$$

15 Finally for the case of a sum of exponential forcings with different time-scales, i.e.  $\sum_{i=1}^n f_i e^{t/\tau_i}$ , we find in a similar way

$$\text{AF} = \frac{\sum_{i=1}^n \frac{1}{1 + \frac{\tau_i}{\tau_s}} f_i e^{t/\tau_i}}{\sum_{i=1}^n f_i e^{t/\tau_i}}.$$

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## Appendix B

### Derivation of the differential equation for the time evolution of the AF

The basis for the derivation of the differential equation for the AF is the general solution of the Eq. (2) for an arbitrary forcing function  $f(t)$ , which is again obtained with the method of “variation of constant”:

$$\Delta C = \int_{-\infty}^t G(t, t') f(t') dt' \text{ with } G(t, t') = e^{-\int_{t'}^t \frac{dt''}{\tau_s(t'')}} \quad (\text{B1})$$

$G(t, t')$  is called the Greens function of the problem. The interpretation of this expression is as follows. The atmospheric perturbation at time  $t$  is given by the sum of “flux pulses” to the atmosphere, each of them damped exponentially in time by  $G(t, t')$  from the moment they have been emitted into the atmosphere. From the definition of AF we then find

$$\text{AF} = \frac{\frac{dC}{dt}}{f} = 1 - \frac{1}{\tau_s} \frac{\Delta C}{f} = 1 - \frac{1}{\tau_s} \frac{\int_{-\infty}^t G(t, t') f(t') dt'}{f}$$

or equivalently

$$(1 - \text{AF}) = \frac{1}{\tau_s} \frac{\int_{-\infty}^t G(t, t') f(t') dt'}{f}$$

The time-derivative of AF is thus

$$\frac{d\text{AF}}{dt} = \frac{1}{f} \frac{df}{dt} - \frac{1}{\tau_s} \frac{\int_{-\infty}^t G(t, t') f(t') dt'}{f} - \frac{1}{\tau_s} \frac{\frac{d}{dt} \int_{-\infty}^t G(t, t') f(t') dt'}{f}$$

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Applying Leibniz's rule

$$\frac{d}{dt} \int_{g(t)}^{h(t)} m(t,s) ds =$$

$$= m(t, h(t)) \frac{dh}{dt} - m(t, g(t)) \frac{dg}{dt} + \int_{g(t)}^{h(t)} \frac{dm(t,s)}{dt} ds$$

to the third term on the right gives

$$\frac{d}{dt} \int_0^t G(t,t') f(t') dt' = G(t,t) f(t) \frac{dt}{dt} + \int_{-\infty}^t \frac{dG(t,t')}{dt} f(t') dt'$$

$$= f(t) - \frac{1}{\tau_s} \int_{-\infty}^t G(t,t') f(t') dt'.$$

Therefore

$$\frac{dAF}{dt} = \left( \frac{1}{f} \frac{df}{dt} + \frac{1}{\tau_s} \frac{d\tau_s}{dt} \right) (1 - AF) - \frac{1}{\tau_s} AF$$

$$= \left( \frac{1}{f} \frac{df}{dt} + \frac{1}{\tau_s} \frac{d\tau_s}{dt} \right) - \left( \frac{1}{\tau_s} + \frac{1}{f} \frac{df}{dt} + \frac{1}{\tau_s} \frac{d\tau_s}{dt} \right) AF.$$

## 10 Appendix C

### Estimation of atmosphere ocean and atmosphere land exchange time constants $\tau_{oc}$ and $\tau_{ld}$

Coupled carbon cycle ocean general circulation models show that there is an approximately linear relationship between the atmospheric perturbation of  $\text{CO}_2$  and ocean carbon uptake (e.g. Gloor et al., 2003). Furthermore we know ocean anthropogenic car-

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bon inventories from ocean surveys (Sabine et al., 2004; Gruber et al., 2009). Based on the approximate linearity

$$F_{at \rightarrow oc}(t) = F_{at \rightarrow oc}(t_{ref}) \frac{pCO_2^{at}(t) - pCO_2^{at}(1765)}{pCO_2^{at}(t_{ref}) - pCO_2^{at}(1765)}$$

and thus

$$5 \quad \tau_{oc} = \frac{pCO_2^{at}(t_{ref}) - pCO_2^{at}(1765)}{F(t_{ref})} = 81.4 \text{ yr}.$$

Here  $t_{ref} = 1995$  and  $F(1995) = 2.2 \text{ PgC yr}^{-1}$  is from Gruber et al. (2009),  $pCO_2(1765) = 276.7 \text{ ppm}$  (Etheridge et al., 1996),  $pCO_2(1995) = 360.9 \text{ ppm}$ , and  $1 \text{ ppm CO}_2 = 2.1276 \text{ PgC}$  for the earth's atmosphere (e.g., Sarmiento et al., 2009). Given  $\tau_s = 37.5 \text{ years}$  (from the main text) and using the relation

$$10 \quad \frac{1}{\tau_s} = \frac{1}{\tau_{ld}} + \frac{1}{\tau_{oc}}$$

from Eq. (2) from the main text we furthermore find  $\tau_{ld} \approx 69.5 \text{ yr}$ .

## Appendix D

### Derivation of a predictive equation for the AF for multiple ocean pools

15 Instead of one differential equation for the evolution of atmospheric  $\Delta C$  we consider a system of ordinary differential equations describing carbon exchange between different volumes of the ocean. The solution of a system of ordinary differential equations is similar to the solution  $\Delta C$  given in Eq. (B1), Appendix B, for Eq. (2) but with Greens function  $G(t, t') = G_{ld}(t, t')G_{oc}(t, t')$  and  $G_{oc}(t, t') = A_0 + \sum_{j=1}^N A_j e^{-\frac{t-t'}{\tau_j}}$ ,  $\sum_{j=0}^N A_j = 1$ . The  
 20 Greens function  $G_{oc}(t, t')$  for the oceans is available from Sarmiento et al. (1992) and

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Maier-Reimer and Hasselmann (1987), calculated using coupled ocean circulation carbon cycle models. The perturbation of atmospheric carbon  $\Delta C$  due to anthropogenic emissions is then given by

$$\Delta C = \int_{-\infty}^t e^{-\frac{t-t'}{\tau_{ld}}} \left( A_0 + \sum_{j=1}^4 A_j e^{-\frac{t-t'}{\tau_j}} \right) f(t') dt'.$$

5 Thus

$$\frac{d\Delta C}{dt} = \sum_{j=0}^4 A_j f(t) +$$

$$- \int_{-\infty}^t e^{-\frac{t-t'}{\tau_{ld}}} \left( \frac{A_0}{\tau_{ld}} + \sum_{j=1}^4 A_j \left( \frac{1}{\tau_{ld}} + \frac{1}{\tau_j} \right) e^{-\frac{t-t'}{\tau_j}} \right) f(t') dt'$$

$$= f(t) - l(t)$$

using  $\sum_{j=0}^4 A_j = 1$  and with

10 
$$l(t) \equiv \int_{-\infty}^t e^{-\frac{t-t'}{\tau_{ld}}} \left( \frac{A_0}{\tau_{ld}} + \sum_{j=1}^4 A_j \left( \frac{1}{\tau_{ld}} + \frac{1}{\tau_j} \right) e^{-\frac{t-t'}{\tau_j}} \right) f(t') dt'.$$

Therefore

$$\frac{dAF}{dt} = \frac{\frac{d\Delta C}{dt}}{f} = \frac{1}{f} \frac{df}{dt} l(t) - \frac{1}{f} \frac{dl}{dt} = \frac{1}{f} \frac{df}{dt} (1 - AF) - \frac{1}{f} \frac{dl}{dt}$$

with

$$\frac{dl}{dt} = \left( \frac{A_0}{\tau_{ld}} + \sum_{j=1}^4 A_j \left( \frac{1}{\tau_{ld}} + \frac{1}{\tau_j} \right) \right) f(t) +$$

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$$-\int_{-\infty}^t e^{-\frac{t-t'}{\tau_{ld}}} \left( \frac{A_0}{\tau_{ld}^2} + \sum_{j=1}^4 A_j \left( \frac{1}{\tau_{ld}} + \frac{1}{\tau_j} \right)^2 e^{-\frac{t-t'}{\tau_j}} \right) f(t') dt'$$

## Appendix E

### T-test for significance of a trend

- 5 For completeness we give here the test statistic for the significance of the slope  $b$  of a regression line  $y = bx + a$  to data  $(x_i, y_i), i = 1, \dots, N$ :

$$t = \frac{b - \beta}{\sqrt{\left( \frac{s^2}{N s_x^2} \right)}}$$

with

$$s^2 = \frac{1}{N-2} \sum_{i=1}^N (y_i - a - bx_i)^2$$

10 
$$s_x^2 = \sum_{i=1}^N (x_i - \bar{x})^2.$$

The  $t$  statistic is distributed as a t-distribution with  $N-2$  degrees of freedom. Because we want to test whether  $b$  differs significantly from 0, we use the statistic for  $\beta = 0$ .

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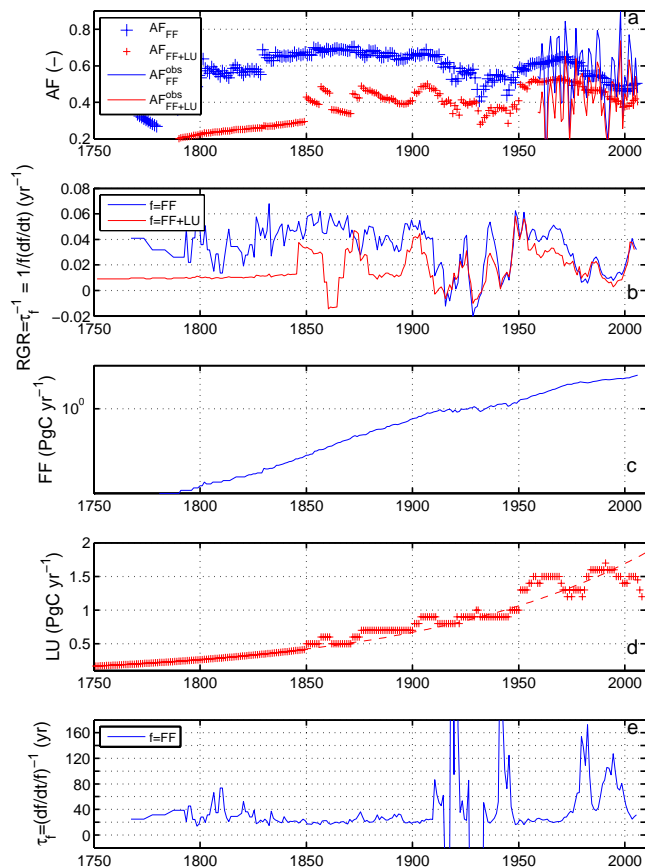
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**Fig. 1.** (a) Model predicted and observed  $AF_{FF}$  and  $AF_{FF+LU}$ , (b) relative growth rate of  $f = FF$  and  $f = FF + LU$  respectively, (c) fossil fuel emissions estimated by Marland (2006), (d) land use flux to the atmosphere estimated by Houghton et al. (2007) due to land use change, and, (e) time scale of relative rate of change of FF and FF + LU respectively.

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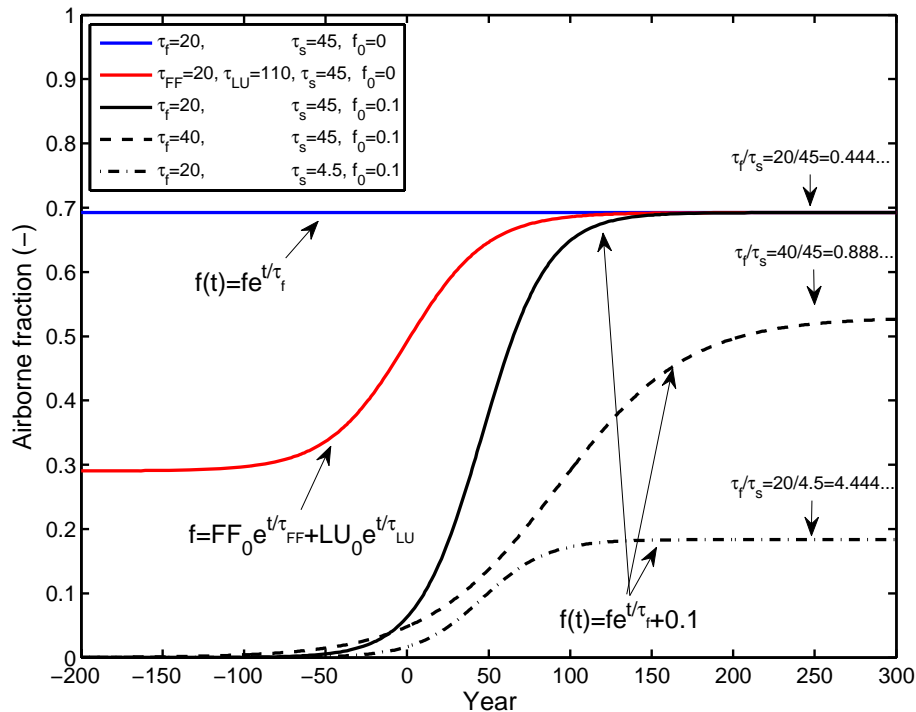
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**Fig. 2.** Predicted air-borne fraction for a range of forcings including purely exponential forcing  $f(t) = fe^{t/\tau_f}$  (blue), mixed exponential and constant forcing  $f(t) = fe^{t/\tau_f} + f_0$  with  $f_0 = 0.1$  (black), and forcing by the sum of two exponential functions,  $f(t) = fe^{t/\tau_f} + f_{LU}e^{t/\tau_{LU}}$  (red).

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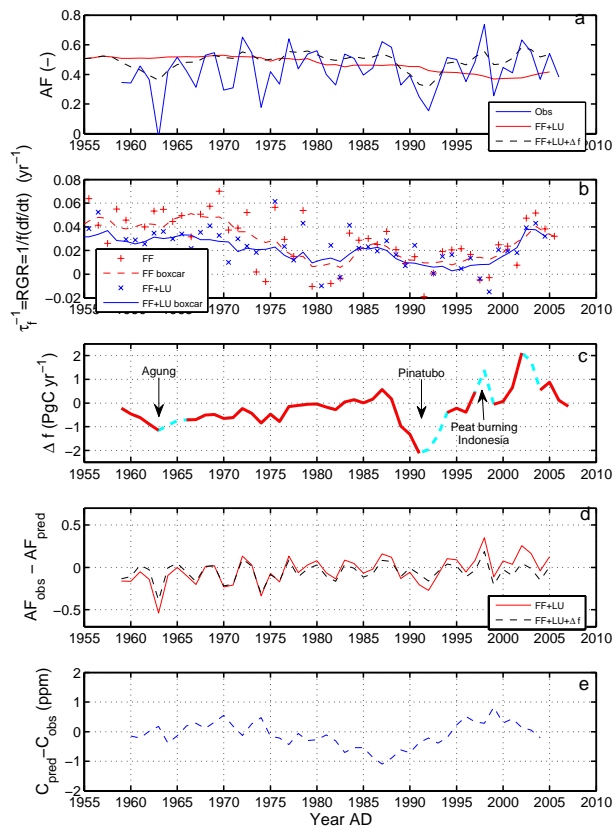
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**Fig. 3.** (a) Model predicted and observed  $AF_{FF}$  and  $AF_{FF+LU}$  for the period from 1950 to 2010, (b) relative growth rate of  $f = FF$  and  $f = FF + LU$  respectively, (c) correction to land use and fossil fuel emissions  $\Delta f$  calculated by minimizing the least square difference between predicted and observed  $AF_{FF+LU}$ , (d) difference between observed and model predicted  $AF_{FF+LU}$  and  $AF_{FF+LU+\Delta f}$ , and (e) difference between predicted and observed atmospheric  $CO_2$ .

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