

1 **Dynamics of Global Atmospheric CO<sub>2</sub> Concentration from 1850 to 2010: a Linear**  
2 **Approximation**

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12

1 **Abstract**

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3 Changes in Earth's temperature have significant impacts on the global carbon cycle, yet  
4 the quantification of such impacts using linear schemes is traditionally deemed difficult.  
5 Here we show that, by incorporating a temperature sensitivity parameter into a simple  
6 linear model, we can satisfactorily characterize the timescale-dependent responses of  
7 atmospheric CO<sub>2</sub> concentration to temperature changes and carbon emissions and  
8 accurately reproduce the history of atmospheric CO<sub>2</sub> between 1850 and 2010. The linear  
9 modeling framework allows us to analytically examine the dynamic characteristics of the  
10 carbon system and associate them with the response times of the carbon reservoirs and  
11 the temperature sensitivity parameter. These results also have important biogeophysical  
12 implications that appear to highlight the intensification of the global carbon cycle. On one  
13 hand, they indicate that the elevated atmospheric CO<sub>2</sub> concentration enhanced land  
14 carbon uptakes at a rate higher than traditionally thought. On the other hand, such  
15 enhanced gross carbon uptakes are partially offset by the increases in global surface  
16 temperatures, which accelerate the release of carbon from the surface reservoirs into the  
17 atmosphere. As a result, the net rate of atmospheric CO<sub>2</sub> sequestration by global land and  
18 oceans has slowed by ~30% since 1960s. We believe the linear modeling framework  
19 outlined in this paper provides a convenient tool to diagnose the observed atmospheric  
20 CO<sub>2</sub> dynamics and monitor their future changes.

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1 **1. Introduction**

2 Anthropogenic CO<sub>2</sub> emissions from fossil-fuel usage and land-use changes have been  
3 almost exponentially increasing since the Industrial Revolution (Fig. 1). Their  
4 accumulation in the atmosphere appears to be changing Earth’s climate (IPCC 2007). The  
5 full strength of anthropogenic CO<sub>2</sub> emissions for changing the climate has not yet been  
6 reached because only 41-45% of the CO<sub>2</sub> emitted between 1850 and 2010 remained in the  
7 atmosphere while the rest was sequestered by lands and oceans (Jones and Cox, 2005;  
8 Canadell et al. 2007; Raupach et al. 2008; Knorr 2009) (Fig. 1). This largely constant  
9 ratio, generally referred to as the “airborne fraction” (denoted as “ $\gamma$ ” in this paper), was  
10 conventionally used to evaluate the efficiency of global carbon sinks (carbon sequestered  
11 by lands and oceans) in assimilating the extra CO<sub>2</sub> from the atmosphere (Jones and Cox,  
12 2005; Canadell et al. 2007). A few recent studies found that the airborne fraction can also  
13 be influenced by other factors, and thus may not be an ideal indicator for monitoring  
14 changes in the carbon sink efficiency (Knorr 2009; Gloor et al. 2010; Frölicher et a.  
15 2013). Nevertheless, the remarkable constancy of the observed airborne fraction provides  
16 important hints for us to examine the dynamic characteristics of the carbon cycle (Gloor  
17 et al. 2010). In particular, it indicates that the responses of atmospheric CO<sub>2</sub>  
18 concentration to the disturbances of anthropogenic CO<sub>2</sub> emissions since 1850s can be  
19 properly approximated by a linear dynamic system with largely constant parameters  
20 (such a linear system is said to be linear time-invariant or LTI; the linear systems  
21 discussed in this paper are assumed to be LTI unless otherwise stated).

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1 | Our reasoning is based on two important observations. First, it is a basic feature of a  
2 | stable linear system that its responses to an exponentially increasing forcing [e.g.,  
3 |  $\exp(\alpha t)$ ,  $\alpha > 0$ ,  $t \geq 0$ ] will approach the input signal with a constant ratio (Naylor and Sell  
4 | 1982). Second, a nonlinear dynamic system can be linearly approximated around a steady  
5 | point within a neighborhood in its state space (Khalil 2001). In the case of the global  
6 | carbon cycle, the existence of such a (quasi) steady state is evident in that the  
7 | atmospheric CO<sub>2</sub> concentration (and the corresponding global climatology) had been  
8 | relatively stable for thousands of years before the industrial era (IPCC 2007). Therefore,  
9 | the observed simple proportional relationship between the atmospheric CO<sub>2</sub>  
10 | concentrations and the increasing CO<sub>2</sub> emissions suggests that recent changes of the  
11 | global carbon cycle are still within the linear neighborhood of the system's last steady  
12 | state.

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14 | There is a rich literature on the application of linear methodology to study the global  
15 | carbon cycle, either to approximate the system's dynamics or diagnose its characteristics  
16 | (e.g., Oeschger and Heimann 1983; Meier-Raimer and Hasselmann 1987; Enting and  
17 | Mansbridge 1987; Wigley 1991; Jarvis et al. 2008; Gloor et al. 2010; Joos et al. 1996,  
18 | 2013). At the heart of some of the most influential methods is the estimation of the  
19 | system's Impulse Response Function (IRF; or more generally the Green's function),  
20 | which describes the time-varying responses of atmospheric CO<sub>2</sub> to a pulse of external  
21 | disturbances, usually anthropogenic carbon emissions. Because the analytical  
22 | determination of IRFs is difficult for complex systems, they were often obtained by  
23 | fitting exponential equations to the numerical experiment results with global carbon-cycle

1 models or their sub-components (Meier-Raimer and Hasselmann 1987; Joos et al. 1996,  
2 2013). Once the IRF is known, the state of atmospheric CO<sub>2</sub> can be conveniently  
3 calculated through linear convolution of the IRF and the records of CO<sub>2</sub> emissions.  
4 Results obtained by such linear approaches well agree with the simulations from the  
5 corresponding global carbon-cycle models unless the disturbances to the system are too  
6 large (Wigley 1991; Li et al. 2009).

7  
8 Although previous studies mostly use IRFs as convenient tools to substitute the  
9 corresponding “parent” models in calculation, the significance of IRFs in diagnosing the  
10 dynamic characteristics of the carbon-cycle system cannot be underestimated. The fact  
11 that IRFs can be represented by a few exponential functions (Meier-Raimer and  
12 Hasselmann 1987) indicates that the dynamic responses of their parent models are largely  
13 captured by a few dominant linear modes (Young 1999) – in other words, the  
14 fundamental dynamic characteristics of these global carbon-cycle models can be learned  
15 from suitable lower-order linear models. For instance, Li et al. (2009) were able to infer  
16 the response (*e*-folding) time constants of the major carbon reservoirs in the carbon-cycle  
17 model of Lenton (2000) by studying its IRF with a fifth-order linear model.

18  
19 Extending the line of thoughts from the literature, this study applies lower-order linear  
20 models to investigate the dynamic characteristics of the global carbon cycle based on  
21 observations. Because the IRF of the real-world system is unknown, we can only treat the  
22 global carbon cycle as a “black box” and use the observed forcing-response relationships  
23 to constrain our models. Nevertheless, the independence from a parent model also gives

1 us more freedom to diagnose some important dynamic modes that have been less  
2 investigated in previous linear models. In particular, global surface temperature has  
3 increased by ~1 °C since the beginning of the 20<sup>th</sup> century (Hansen et al. 1999; Brohan et  
4 al. 2006). Given the tight coupling between temperature and the carbon cycle (Keeling et  
5 al. 1995; Joos et al. 1999, 2001; Lenton 2000; Rafelski et al. 2009), the warming alone  
6 may release a large amount of CO<sub>2</sub> from the land and the oceans into the atmosphere,  
7 redistributing carbon among these reservoirs. Previous studies have noticed that the  
8 effects of temperature on atmospheric CO<sub>2</sub> vary at different time scales, ranging from 1-2  
9 ppm °C<sup>-1</sup> at the scale of years to 10-20 ppm °C<sup>-1</sup> over millennium or centuries (see the  
10 literature review by Woodwell et al. 1998). However, such effects are traditionally  
11 deemed difficult to quantify by simple schemes (Scheffer et al. 2006). Here we show that,  
12 by using a simple sensitivity parameter to represent the effect of temperature in our linear  
13 model, we can satisfactorily characterize the dynamic responses of atmospheric CO<sub>2</sub>  
14 concentration to temperature changes (and carbon emissions) as reported in the literature  
15 while accurately reproducing the history of CO<sub>2</sub> in the atmosphere in the past 160 years  
16 (see below).

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18 A practical factor to decide in developing a diagnostic model for the global carbon cycle  
19 is the complexity of the linear tool itself. This may not represent a serious difficulty in the  
20 forward model construction and analysis, where well-established mathematical tools are  
21 at our disposal (see the example in the Appendix). For the inverse problem of model  
22 identification, on the other hand, it is the resolution of available observations that  
23 essentially determines the number of independent system parameters that can be reliably

1 | retrieved. In this study, we decided to demonstrate our analytical framework by a simple  
2 | two-box model that represents carbon exchanges between the atmosphere and the surface  
3 | (i.e. land and ocean) reservoirs. This decision is based on multiple considerations besides  
4 | the constraints of model identification, which include that, for instance, the analysis of a  
5 | two-box model involves only simple mathematical techniques but render clear physical  
6 | pictures of the problem under investigation. Though such a “toy” model may sit at the  
7 | lowest rank on the hierarchy of global carbon-cycle models (Enting 1987), new and  
8 | important characteristics of the atmospheric CO<sub>2</sub> dynamics can still be learned from it.  
9 | Furthermore, the use of a simple model by no means implies the compromise of scientific  
10 | rigor of our findings, which are verified in a generalized linear model framework as  
11 | described in the Appendix.

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13 | Throughout the analysis we also compare the results obtained from the two-box model to  
14 | those from the more advanced Bern model (Siegenthaler and Joos 1992; Enting et al.  
15 | 1994; IPCC 1996, 2001). The Bern model couples the atmosphere with a process-based  
16 | ocean biogeochemical scheme (Siegenthaler and Joos 1992; Shaffer and Sarmiento 1995;  
17 | Joos et al. 1999) and a multi-component terrestrial biosphere module (Siegenthaler and  
18 | Oeschger 1987). The original Bern model does not consider the effects of changing  
19 | global temperatures on terrestrial ecosystem respiration, which however plays an  
20 | important role in regulating the variability of the global carbon cycle at interannual to  
21 | multi-decadal time scales (Wang et al. 2013; Rafelski et al. 2009). Therefore, we revised  
22 | the Bern model to account for temperature’s effects on terrestrial ecosystem respiration  
23 | and recalibrate the model subsequently (see the Appendix for details). The global carbon-

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1 cycle processes described in the Bern model help us diagnose the biogeophysical  
2 mechanisms underlying the characteristics of the atmospheric CO<sub>2</sub> dynamics identified  
3 with our simple linear model.

4

## 5 **2. Datasets**

6 Annual atmospheric CO<sub>2</sub> concentration data from 1850 to 1960 are based on the ice core  
7 CO<sub>2</sub> records from Law Dome, Antarctica (Etheridge et al. 1996) and those between 1960  
8 and 2010 are compiled from NOAA Earth System Research Laboratory (ESRL) (Keeling  
9 et al. 1995; Conway et al. 1994). We merged the data following the approach described in  
10 Le Quéré et al. (2009) and calculated annual CO<sub>2</sub> growth rate as the first-order difference  
11 of the yearly CO<sub>2</sub> concentrations. Long-term records of anthropogenic CO<sub>2</sub> emissions  
12 from fossil fuel burning and cement production are compiled by Boden et al. (2011) and  
13 those of land-use changes are from Houghton (2003), both downloaded from the Carbon  
14 Dioxide Information Analysis Center at Oak Ridge National Laboratory, TN, USA  
15 (<http://cdiac.ornl.gov>). Two sets of monthly surface temperature data are used, including  
16 GISTEMP from NASA Goddard Institute for Space Studies (Hansen et al. 1999) and the  
17 CRU-NCEP climate dataset (Sitch et al. 2008; Le Quéré et al. 2009), available from 1901  
18 to the present with spatial resolutions of 0.5×0.5 (CRU-NCEP) or 1×1 (GISTEMP)  
19 degrees. Monthly time series of temperature are aggregated globally and over the tropics  
20 (24°N-24°S), and smoothed with a 12-month running window to convert the monthly data  
21 to annual values. We calculated temperature anomalies relative to their 1901 to 1920  
22 annual mean and assumed the 20-years mean temperature to be representative of  
23 temperature climatologies between 1850 and 1900. This assumption is reasonable as



1 suggested by analysis of other long-term coarse-resolution temperature datasets (Jones et  
2 al. 2003; Brohan et al. 2006).

3

### 4 3. Derivation of the Two-Box Model

5 This study considers only the “fast” carbon flows between the atmosphere and the surface  
6 at time scales within hundreds of years (IPCC 2001). In the two-box approach discussed  
7 below the world’s land and oceans are treated as one combined carbon reservoir (“box”).  
8 A generalized treatment of the surface carbon reservoirs by individuals is presented in the  
9 Appendix.

10

11 Based on our linearization assumption, we describe the dynamics of the two-box carbon  
12 system using the following equations:

$$13 \quad \dot{A}' = -\alpha_A \cdot A' + \alpha_S \cdot S' + \beta_T \cdot T' + \dot{E}' \quad (1a)$$

$$14 \quad \dot{S}' = +\alpha_A \cdot A' - \alpha_S \cdot S' - \beta_T \cdot T' \quad (1b)$$

15 where  $A$  and  $S$  denote carbon storages in the atmosphere and the surface reservoirs,  
16 respectively, and  $E$  is the accumulated anthropogenic  $\text{CO}_2$  emissions since the industrial  
17 era. The three variables can be measured by the same unit of parts per million by volume  
18 (1 ppm =  $\sim 2.13 \times 10^9$  metric-ton carbon or GtC). The prime symbol (e.g., “ $E'$ ”) indicates  
19 that changes in a variable relative to its preindustrial steady-state level. The preindustrial  
20 emissions are assumed negligible so that  $E' = E$ . The dot accent (e.g., “ $\dot{E}'$ ”) indicates the  
21 first-order derivative with regard to time, such that  $\dot{E}'$  represents the annual rate of  $\text{CO}_2$   
22 emissions ( $\text{ppm yr}^{-1}$ ). The positive constant parameters  $\alpha_A$  and  $\alpha_S$  ( $\text{yr}^{-1}$ ) describe the  
23 decaying rates of corresponding carbon anomalies. Their reciprocals (i.e.,  $\tau_A = 1/\alpha_A$ ,

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1  $\tau_S = 1/\alpha_S$ ) are often referred to as the response time of the carbon reservoirs (IPCC  
2 2001).  $T$  ( $^{\circ}\text{C}$ ) denotes indices of global (or large-scale) surface temperatures and the  
3 coefficient  $\beta_T$  ( $\text{ppm yr}^{-1} \text{ } ^{\circ}\text{C}^{-1}$ ) represents the sensitivity of atmospheric  $\text{CO}_2$  growth rate to  
4 temperature changes. The term  $\beta_T T'$  thus indicates the impacts of temperature increases  
5 on the global carbon cycle, which release  $\text{CO}_2$  from the surface reservoirs to the  
6 atmosphere. In this study we have assumed  $\beta_T$  to be a constant. This assumption is  
7 justified later in the paper.

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8  
9 The physical meaning of Eq. 1(a, b) is clear: carbon outflows from the atmosphere,  $\alpha_A A'$ ,  
10 are inflows to the surface (e.g., through photosynthesis in green vegetation and the  
11 dissolution of  $\text{CO}_2$  in the surface water) while carbon outflows from the surface,  $\alpha_S S'$ ,  
12 (e.g., through respiration and the outgassing of the dissolved  $\text{CO}_2$ ) are the inflows to the  
13 atmosphere. The effects of temperature changes,  $\beta_T T'$ , revise the relative carbon balance  
14 between the atmosphere and the surface reservoirs. Human emissions of  $\text{CO}_2$ , on the  
15 other hand, represent an “external” source of  $\text{CO}_2$  to the system by rapidly releasing  
16 carbon (e.g., fossil fuel burning) from reservoirs that were formed over millions of years  
17 and by permanently altering the structure of land surface carbon pools (e.g., land-  
18 cover/land-use changes).

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19  
20 Because mass (carbon) is conserved in the two-box model, Eqs. (1a) and (1b) are not  
21 independent. Adding the two equations together leads to

$$\dot{A}' + \dot{S}' = \dot{E}'$$

22  
23 or

1 
$$A' + S' = E'. \tag{1c}$$

2 Eq. (1c) simply states that the anthropogenically emitted CO<sub>2</sub> either resides in the  
3 atmosphere or in the surface reservoirs (i.e., the land and the oceans). Substituting this  
4 relationship into Eq. (1a) to replace S', we obtain

5 
$$\dot{A}' + (\alpha_A + \alpha_S) \cdot A' = \beta_T \cdot T' + \alpha_S \cdot E' + \dot{E}'. \tag{2a}$$

6 Therefore, the dynamics of atmospheric CO<sub>2</sub> represented by the two-box model is  
7 determined by an ordinary differential equation of A' under the disturbances of  
8 anthropogenic emissions (E' and  $\dot{E}'$ ) and the changing climate (T').

9

#### 10 4. Model Determination and Evaluation

11 We want to determine the parameters of Eq. (2) with observational records of A', T', and  
12 E', and evaluate how well the model captures the observed atmospheric CO<sub>2</sub> dynamics.

13 In order to construct a regression model from Eq. (2), we rearrange the equation as  
14 follows:

15 
$$\dot{E}' - \dot{A}' = (\alpha_A + \alpha_S) \cdot A' - \alpha_S \cdot E' - \beta_T \cdot T', \tag{2b}$$

16 where  $\dot{E}' - \dot{A}'$  represents the strength of annual carbon sinks. However, because  
17  $A' \approx \gamma \cdot E'$  (Fig. 1), where  $\gamma$  is the airborne fraction, the “collinearity” between the two  
18 regressors prevents us from determining the coefficients associated with them separately  
19 (Chatterjee and Hadi 2006). Indeed, substituting the airborne-fraction relationship into  
20 Eq. (2b) leads to

21 
$$\dot{E}' - \dot{A}' = [\alpha_A - (1/\gamma - 1)\alpha_S] \cdot A' - \beta_T \cdot T', \tag{2c}$$

22 which implies that only a combination of  $\alpha_A$  and  $\alpha_S$  can be estimated from the  
23 observations, and the estimation of  $\beta_T$  also needs some special care (see below).

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2 We may reach the above argument from another perspective. Fig. (1) shows that the  
3 anthropogenic CO<sub>2</sub> emissions ( $E'$  and  $\dot{E}'$ ) can be approximated by exponential functions.  
4 Let  $E' = \exp(\alpha_E t)$ , and it follows that  $\dot{E}' = \alpha_E \cdot \exp(\alpha_E t)$ . By applying the airborne  
5 fraction relationship ( $A' \approx \gamma \cdot E'$  and  $\dot{A}' \approx \gamma \cdot \dot{E}'$ ) and neglecting the influence of  
6 temperature ( $\beta_T T'$ ) for now, we obtain

$$7 \quad \gamma \cdot [\alpha_E + (\alpha_A + \alpha_S)] \cdot \exp(\alpha_E \cdot t) \approx (\alpha_S + \alpha_E) \cdot \exp(\alpha_E \cdot t),$$

8 or

$$9 \quad \alpha_A - (1/\gamma - 1) \cdot \alpha_S \approx (1/\gamma - 1) \cdot \alpha_E. \quad (2d)$$

10 The term on the left-hand side of Eq. (2d) is the same as the (regression) coefficient  
11 associated with the  $A'$  in Eq. (2c). Eq. (2d) suggests that this coefficient is closely related  
12 to the exponential disturbances ( $\alpha_E$ ) to the system. Because  $\alpha_E > 0$  and  $\gamma$  is about 0.41-  
13 0.45 (Fig. 1), it follows that  $(1/\gamma - 1) > 1$  and  $\alpha_A > \alpha_S$  (or  $\tau_A < \tau_S$ ).

14

15 The above analysis indicates that additional information is required to resolve  $\alpha_A$  and  $\alpha_S$   
16 from the regression results of Eq. (2c). One source of such information comes from  
17 previous observation-based studies. For instance, by comparing the carbon isotope ratios  
18 in wood and in marine material, Revelle and Suess (1957) have long suggested that the  
19 response time ( $\tau_A$ ) of atmospheric CO<sub>2</sub> is on the order of 10 years. We also extract  
20 information from process-based model studies. Because the initial decaying rate of the  
21 IRF of a global carbon-cycle model is mainly determined by  $\alpha_A$  (or  $\tau_A$ ; see the proof in  
22 the next section and in the Appendix), applying this result to analyze the ensemble IRFs  
23 reported in Joos et al. (2013) suggests  $\tau_A$  to be ~14 years. We choose  $\tau_A$  to be 12 years

1 ( $\alpha_A \approx 0.083 \text{ yr}^{-1}$ ) so that the IRF of our linear model closely matches with the Bern  
2 model during the initial decaying stage (see the next section). We subsequently estimate  
3  $\tau_S$  to be  $\sim 34$  years ( $\alpha_S \approx 0.029 \text{ yr}^{-1}$ ).

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4  
5 The estimation of the  $\beta_T$  parameter in Eq. (2) requires the choice of a large-scale  
6 temperature index that is representative of climate change and closely related with global  
7 carbon cycle. Previous studies showed that the land surface air temperature in the tropics  
8 (24S-24N) are most strongly coupled with interannual variations in the growth rate of  
9 atmospheric CO<sub>2</sub> by a sensitivity ( $\beta_T$ ) of  $\sim 1.64 \text{ ppm yr}^{-1} \text{ }^\circ\text{C}^{-1}$  (Adams and Piovesan 2005;  
10 Wang et al. 2013). Here we found that the same temperature-CO<sub>2</sub> coupling also operate at  
11 longer time scales. Indeed, because the system is linear, variations in  $A'$ ,  $T'$ ,  $E'$ , and their  
12 derivatives over different time scales must satisfy Eq. (2) separately. Because the  
13 interannual variations (“IAV”) in the emissions (both  $\dot{E}'$  and  $E'$ ) and the atmospheric CO<sub>2</sub>  
14 concentration ( $A'$ ) are relatively small (Fig. 1), neglecting them in Eq. (2) leads to

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$$\dot{A}'_{IAV} \approx \beta_T \cdot T'_{IAV}, \quad (2e)$$

16 which is the same linear relationship as previous reported (Wang et al. 2013).

17  
18 There is another practical reason that we use the  $\beta_T$  estimated from Wang et al. (2013) in  
19 this study. Because the long-term increases in global temperature ( $T'$ ) are mainly induced  
20 by the growing CO<sub>2</sub> concentrations in the atmosphere ( $A'$ ), the two variables are indeed  
21 significantly correlated ( $r \approx 0.9$ , with IAV in them removed). Therefore, estimating  $\beta_T$   
22 directly from Eq. (2c) is inevitably subject to the influence of the collinearity between  $A'$   
23 and  $T'$  (Enting 2010). On the other hand, the short-term variations (i.e., IAV) of global

1 temperature are dominated by the natural variability of the climate system (e.g., the El  
2 Niño-Southern Oscillations). Therefore, we expect the  $\beta_T$  estimated with Eq. (2e) in  
3 Wang et al. (2013) to have less uncertainties.

4  
5 With the model parameters determined, we use the two-box model to simulate the  
6 changes of atmospheric CO<sub>2</sub> concentration between 1850 and 2010 from historical  
7 records of temperature and CO<sub>2</sub> emissions (Fig. 2). The simulated results follow the  
8 evolution of the observed atmospheric CO<sub>2</sub> concentration to a high degree of accuracy,  
9 capturing more than 96% of the variability (i.e.,  $r^2 > 0.96$ ) of the latter (Fig. 2). The  
10 standard deviations ( $\sigma$ ) of the differences between simulated values and those measured  
11 accurately since 1960 are ~0.9 ppm for the atmospheric CO<sub>2</sub> concentration and ~0.4 ppm  
12 for its growth rate, respectively (Fig. 2). These results are highly comparable to those  
13 simulated with the revised Bern model (Fig. 2) or other sophisticated climate-carbon  
14 models reported in the literature (e.g., Joos et al. 1999; Lenton 2000; Friedlinstein et al.  
15 2006), strongly supporting our argument that the atmospheric CO<sub>2</sub> dynamics in the past  
16 one and half centuries can be properly approximated with linear models.

## 18 5. Disturbance-Response Functions

19 We first check the model's responses to an impulse disturbance of anthropogenic CO<sub>2</sub>  
20 emissions. Shown in Fig. 3, the initial atmospheric CO<sub>2</sub> anomaly decays relatively fast,  
21 as 60-70 % of the emitted CO<sub>2</sub> is absorbed by the surface reservoirs within 20 years of  
22 the disturbance. However, the rate of carbon assimilation by the land and the oceans  
23 significantly slows down in the following decades and eventually becomes neutral as the

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... [4]

1 system approaches steady-state. In the end, 15-25% of the simulated CO<sub>2</sub> anomaly will  
2 likely stay in the atmosphere for thousands of years (Fig. 3). These results are consistent  
3 with the findings from fully coupled climate-carbon models (Cao et al. 2009; Archer et  
4 al. 2009; Joos et al. 2013).

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5  
6 The JRF of the linear box models can be analytically characterized. For the two-box  
7 model of Eq. (2), when the system approaches a (new) steady state after the disturbance,  
8 all the time derivatives ( $\dot{E}'$  and  $\dot{A}'$ ) will be zero. Assuming that temperature does not  
9 change during the process, we easily obtain the steady state of  $A'$  as

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10 
$$A' = \frac{\alpha_S}{\alpha_A + \alpha_S} \cdot E' = \frac{\tau_A}{\tau_A + \tau_S} \cdot E', \quad (3a)$$

11 or more generally

12 
$$\frac{A'}{\tau_A} = \frac{S'}{\tau_S}, \quad (3b)$$

13 where the mass-conservation relationship represented by Eq. (1d) is used in the  
14 derivation. Therefore, the extra CO<sub>2</sub> added to the “fast” carbon cycle by anthropogenic  
15 emissions will be partitioned between the atmosphere and the surface corresponding to  
16 the response times ( $\tau$ ) of the reservoirs, respectively. Because  $\tau_S > \tau_A$  (see Eq. (2d) and  
17 the related discussions), a majority of the emitted CO<sub>2</sub> will eventually be absorbed by the  
18 surface carbon reservoirs (Fig. 3). In other words, the long-term fate of the CO<sub>2</sub> emitted  
19 into the atmosphere is largely determined by the response times of the surface reservoirs  
20 (Revelle and Suess 1957).

21

1 The rates at which the atmospheric CO<sub>2</sub> anomaly decays are determined by the solutions  
2 (i.e., eigenvalues) to the characteristic equation of the system. For a two-box system like  
3 Eq. (2a), the problem is particularly simple because the only non-zero eigenvalue ( $\lambda$ ) is

$$4 \quad \lambda = \alpha_A + \alpha_S, \quad (4)$$

5 and the solution of Eq. (2) is therefore

$$6 \quad A' = \frac{\alpha_A}{\alpha_A + \alpha_S} \exp[-(\alpha_A + \alpha_S) \cdot t] + \frac{\alpha_S}{\alpha_A + \alpha_S}. \quad (5a)$$

7 A helpful observation of Eq. (5) is that, when  $t \ll 1/(\alpha_A + \alpha_S)$ , the solution can be  
8 approximated by

$$9 \quad A' \approx \frac{\alpha_A}{\alpha_A + \alpha_S} [1 - (\alpha_A + \alpha_S) \cdot t] + \frac{\alpha_S}{\alpha_A + \alpha_S} = 1 - \alpha_A \cdot t \approx \exp(-\alpha_A \cdot t). \quad (5b)$$

10 That is,  $A'$  initially decays at a maximum rate of  $\alpha_A$  as if the capacity of the surface  
11 carbon reservoir were unlimited (i.e.,  $\alpha_S=0$ ). This result is also valid for general cases  
12 (see the Appendix).

13

14 Next we consider the system's responses to disturbances induced by changes in surface  
15 temperatures. Unlike anthropogenic CO<sub>2</sub> emissions, changes in temperature do not add  
16 additional CO<sub>2</sub> to the "fast" carbon cycle but only re-distribute carbon between the  
17 atmosphere and the surface (Eqs. 1a and 1b), and so the system will recover to its initial  
18 steady state once the temperature anomaly is removed. However, increases in  
19 temperature are persistent under climate-change scenarios. Therefore, we examine the  
20 responses of atmospheric CO<sub>2</sub> to a step change in temperature, which is determined from  
21 Eq. (2) as:

$$22 \quad A' = \beta_T / (\alpha_A + \alpha_S) \cdot T'. \quad (6a)$$

23 Because  $\alpha_A > \alpha_S$ , for rough estimates we can also use



1  $A' \approx \beta_T / \alpha_A \cdot T'$  (6b)

2 Based on the estimated model parameters, therefore, atmospheric CO<sub>2</sub> rises by ~15 ppm  
3 for an increase of 1 °C in temperature within a few decades (Fig. 3). This long-term  
4 temperature sensitivity of atmospheric CO<sub>2</sub> is consistent with the estimate inferred from  
5 the (reconstructed) temperature and atmospheric CO<sub>2</sub> records during the Little Ice Age  
6 (~20 ppm °C<sup>-1</sup>; see Woodwell et al. 1998).

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8 The relationships represented by Eqs. (3b), (5b), and (6b) can be generalized to higher-  
9 order systems (see the Appendix), providing a convenient way to characterize the  
10 models' disturbance-response functions without fully solving the system equations.  
11 However, the uncertainties associated with these results – especially the long-term  
12 responses of atmospheric CO<sub>2</sub> – need to be emphasized. One key source of the  
13 uncertainties is that model's parameters are not fully determined by the observations of  
14 the global climate-carbon system. As discussed in Section 4, the estimation of the model  
15 parameter  $\alpha_S$  depends on the choice of  $\alpha_A$ , which is only loosely constrained by the prior  
16 knowledge. The analysis in the Appendix indicates that this situation only worsens in  
17 higher-order (N-box) systems as the number of system parameters increase at the order of  
18 N<sup>2</sup> (also see Joos et al. 1996). It is possible for us to choose another pair of  $\alpha_A$  and  $\alpha_S$  or a  
19 higher-order linear model so that the derived disturbance response functions better  
20 approximate those of the Bern model. However, tuning the model in this fashion has only  
21 cosmetic effects to the results and does not reduce the associated uncertainties. In  
22 addition, in reality the climate system and the global carbon cycle are not independent but  
23 tightly coupled. Therefore, a comprehensive assessment of the long-term fate of

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1 anthropogenic CO<sub>2</sub> emissions in the atmosphere must account for the effects of the  
2 associated changes in global temperature, which is beyond the scope of this study.

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#### 4 6. Biogeophysical Implications

5 The above analysis suggests that the appropriate representation of temperature's effects  
6 on the carbon cycle in our linear model helps improve the model's accuracy in  
7 approximating the observed dynamics of the atmospheric CO<sub>2</sub> across multiple time  
8 scales. To illustrate, we further rearrange Eq. (2c) to obtain

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$$9 \quad \dot{E}' - \dot{A}' + \beta_T \cdot T' = [\alpha_A - (1/\gamma - 1)\alpha_S] \cdot A'. \quad (7)$$

10 On the left-hand side of the equation, the term “ $\dot{E}' - \dot{A}'$ ” is usually used to measure the  
11 *net* strength of annual global carbon sinks. However, because the warming temperature  
12 also releases carbon on from the surface into the atmosphere ( $\beta_T \cdot T'$ ), this extra source of  
13 CO<sub>2</sub> has to be absorbed by the global carbon sinks. By accounting for the effects of  
14 temperature changes, the term “ $\dot{E}' - \dot{A}' + \beta_T \cdot T'$ ” thus define a *gross* global carbon  
15 sinks.

16  
17 Examining Eq. (7) with the observational data shows that both the net and the gross  
18 carbon sinks have been steadily increasing in response to the rising atmospheric CO<sub>2</sub>  
19 concentration in the past 160 years, reaching  $\sim 2.5$  ppm yr<sup>-1</sup> and  $\sim 4.0$  ppm yr<sup>-1</sup>  
20 respectively in 2010 (Fig. 4). The gross carbon sinks have a nearly direct linear  
21 relationship (with a constant slope  $\sim 0.04$  yr<sup>-1</sup>;  $r=0.98$ ) with the atmospheric CO<sub>2</sub>  
22 concentrations throughout the entire data period. In comparison, the relationship between  
23 the apparent carbon sinks and the CO<sub>2</sub> concentrations is slightly nonlinear, with its slope

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1 decreasing from  $\sim 0.03 \text{ yr}^{-1}$  in 1960 to  $\sim 0.02 \text{ yr}^{-1}$  in 2010. Therefore, our linear  
2 approximation approach would not be able to achieve the same high accuracy if  
3 temperature's effects on the carbon cycle were not correctly represented. Note that the  
4 slope of these linear relationships, (i.e.,  $\alpha_A - (1/\gamma - 1)\alpha_S$ ), is sometimes interpreted as  
5 the efficiency of surface carbon reservoirs in sequestering annual CO<sub>2</sub> emissions (Gloor  
6 et al. 2010; Raupach et al. 2014). Since this coefficient is influenced by the AF factor ( $\gamma$ ),  
7 it is not an intrinsic characteristic of the carbon-cycle system. Therefore, the "sink  
8 efficiency" interpretation of the coefficient is only meaningful when  $\gamma$  is relatively  
9 constant. Nevertheless, Fig. 4 shows that although the *gross* carbon-sequestration rates of  
10 the surface reservoirs changed little, the *net* "efficiency" of the system has slowed by  
11  $\sim 30\%$  in the past five decades. This finding is essentially the same as reported in Raupach  
12 et al. (2014) but our analysis emphasizes that this declining carbon sequestration rate,  
13 mainly reflects the impacts of climate changes on the global carbon cycle.

14  
15 The biogeophysical implication of the parameter  $\beta_T$  needs further discussion. Our  
16 previous analysis (Wang et al. 2013) suggests that this parameter mainly reflects the  
17 temperature sensitivity of respiration of land-surface carbon pools (biomass and soil  
18 carbon). This explanation is supported by the simulations of the Bern model in this study,  
19 in which terrestrial carbon sinks have much stronger responses to temperature changes  
20 than the ocean counterpart (not shown). Furthermore, both our simulations and those  
21 from the literature (e.g., Canadell et al. 2007; Le Quéré et al. 2009) indicate that the total  
22 carbon storage in the land-surface reservoirs remains largely stable between 1850 and  
23 2010, a necessary condition for  $\beta_T$  to be constant. For instance, because terrestrial carbon

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**Deleted:** Because the efficiency of sea-air carbon exchanges is constrained by the buffering effects of the surface biogeochemistry (Revelle and Suess 1957), ocean carbon uptakes simulated by the Bern model account for  $\sim 50\%$  of the *net* global sinks, while the other 50% is contributed by land processes, consistent with the literature [Canadell et al. 2007; Le Quéré et al. 2009].

1 uptake accounts for 50-60% of the global net sinks in our simulations, the accumulated  
2 terrestrial net carbon sinks are about 71-85 ppm in 2010, representing a 7-8% increase in  
3 the total terrestrial carbon storage (~1040 ppm as of 1850). At the same time, the  
4 accumulated terrestrial carbon losses through land-use changes are about 74 ppm in 2010  
5 based on the dataset of Houghton (2003). These results suggest that the net changes in the  
6 total terrestrial biomass and soil carbon are (relatively) small during the past 160 year,  
7 providing further justification for our linear modeling approach.

8  
9 Finally, our analysis suggests that the increasing atmospheric CO<sub>2</sub> concentration must  
10 have promoted carbon assimilation by the terrestrial biosphere (Ballantyne et al. 2012),  
11 most likely through the CO<sub>2</sub> fertilization effect (Körner and Arnone 1992; Oechel et al.  
12 1994; Long et al. 1991, 2004) and the associated ecological changes (Keenan et al. 2013;  
13 Graven et al. 2013). Indeed, because the surface warming rapidly releases a proportion of  
14 the assimilated carbon back to the atmosphere (Fig. 4) (Piao et al. 2008; Wang et al.  
15 2013), the increased turnover rate may have obscured the evaluation of the magnitude of  
16 the CO<sub>2</sub> fertilization effects, which we found in calibrating the Bern model (see the  
17 Appendix). In other words, the gross CO<sub>2</sub> fertilization effect of terrestrial vegetation is  
18 likely higher than previously thought (Schimel et al. 2014).

## 20 7. Conclusions

21 We demonstrate in this paper that the observed dynamics of the global atmospheric CO<sub>2</sub>  
22 concentration from 1850 to 2010 can be properly approximated as a linear system. In

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**Deleted:** the gross CO<sub>2</sub> fertilization effects of terrestrial vegetation may be higher than previously thought:

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1 particular, we derived a simple box model to describe carbon exchanges between the  
2 atmosphere and the surface carbon reservoirs under the disturbances of anthropogenic  
3 CO<sub>2</sub> emissions as well as global temperature changes. We show that, with a few  
4 appropriately retrieved parameters, the model can successfully simulate the observed  
5 changes and variations of the atmospheric CO<sub>2</sub> concentration and its first-order derivative  
6 (i.e., CO<sub>2</sub> growth rate) across interannual to multi-decadal time scales. The results are  
7 highly comparable to those obtained with more sophisticated models in the literature,  
8 confirming that the simple linear model is capable in capturing the main features of  
9 atmospheric CO<sub>2</sub> dynamics in the past one and half centuries.

10

11 A distinct advantage of our linear modeling framework is that it allows us to analytically,  
12 and thus most directly, examine the dynamic characteristics of the (modeled) carbon-  
13 cycle system. Our analyses indicate that many of such characteristics are closely  
14 associated with the response times of the atmosphere and surface carbon reservoirs. For  
15 instance, the initial decaying rate of an impulse of CO<sub>2</sub> emitted into the atmosphere is  
16 mainly influenced by the response time of the atmosphere, but the proportion of the extra  
17 CO<sub>2</sub> that stays in the atmosphere at long-term time scales is determined by the ratio  
18 between the response times of the atmosphere and the surface reservoirs. Unfortunately,  
19 the collinearity exhibited by the observed time series of CO<sub>2</sub> emissions and atmospheric  
20 CO<sub>2</sub> concentrations has obscured the determination of the response times for individual  
21 surface reservoirs, inducing uncertainties of the estimated long-term responses of the  
22 global carbon system. In other words, although the steady CO<sub>2</sub> airborne fraction allows

1 us to represent the carbon-system dynamics with a simple model, it prevents us to resolve  
2 further details of the surface carbon reservoirs.

3

4 Our model results also have important biogeophysical implications. They highlight that  
5 the responses of the global carbon cycle to recent anthropogenic and climatic  
6 disturbances are still within the resilience zone of the system, such that annual (gross)  
7 terrestrial and ocean carbon sinks linearly increases with the atmospheric CO<sub>2</sub> levels. On  
8 one hand, the elevated atmospheric CO<sub>2</sub> concentration must have enhanced land carbon  
9 uptakes through the “fertilization” effects and the associated ecological changes. On the  
10 other hand, ~~the enhanced gross carbon uptakes are partially offset by the increases in~~  
11 global surface temperatures, which accelerate the release of carbon from the surface  
12 reservoirs into the atmosphere. As a result, the “net” efficiency of global land and oceans  
13 in sequestering atmospheric CO<sub>2</sub> may have slowed by ~30% since 1960s, although the  
14 airborne fraction of CO<sub>2</sub> emissions remains largely constant.

15

16 Finally and importantly, we emphasize that the linear approximation of the global carbon  
17 cycle discussed in this paper is conditioned on the pre-industrial (quasi) steady state of  
18 the system. The global climate-carbon system is clearly nonlinear beyond this scope  
19 (Archer et al. 2009), which can establish different steady states over glacial/interglacial  
20 time scales (Sigman and Boyle 2000). A major concern stemming from climate change is  
21 that, because the post-industrial anthropogenic disturbances on the global carbon cycle  
22 are so strong and rapid, they may abruptly alter the pace at which the natural climate-  
23 carbon system evolves and drive the system into a different state at a drastically

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1 accelerated rate (IPCC 2001). Our results clearly indicate that the rising atmospheric CO<sub>2</sub>  
2 concentrations and the associated increases in global temperature have significantly  
3 intensified the global carbon cycle in the past one and half centuries. Although such  
4 intensification of the carbon system seems to be within the linear zone as of now, its  
5 resilience may be weakened, or lost, in the future. As the anthropogenic CO<sub>2</sub> emissions  
6 continue to increase and the global temperature continues to warm, scientists generally  
7 expect surface – in particular, terrestrial – carbon reservoirs to saturate and their CO<sub>2</sub>  
8 sequestration efficiency to decrease, such that the responses of the global carbon cycle to  
9 the anthropogenic disturbances will eventually deviate from their original path. With this  
10 concern regarded, the simple linear model developed in this study may serve as a  
11 convenient tool to monitor the early signs when the natural carbon system is pushed away  
12 (by anthropogenic disturbances) from its linear zone.

13

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## 1 **Appendix**

### 2 **A.1 Calibrations of the Bern carbon-cycle model**

3 The Bern model is a coupled global carbon-cycle box model (Siegenthaler and Joos  
4 1992; Enting et al. 1994) that was used in previous IPCC Assessment Reports to study  
5 changes in atmospheric CO<sub>2</sub> concentration under different emission scenarios (IPCC  
6 1996, 2001). It couples the High-Latitude Exchange/Interior Diffusion-Advection  
7 (HILDA) ocean biogeochemical model (Siegenthaler and Joos 1992; Shaffer and  
8 Sarmiento 1995; Joos et al. 1999) with an atmosphere layer and a multi-component  
9 terrestrial biosphere model (Siegenthaler and Oeschger 1987). The HILDA model  
10 describes ocean biogeochemical cycling through two well-mixed surface layers in low  
11 and high latitudes, a well-mixed deep ocean in the high latitude and a dissipative interior  
12 ocean in the low latitude. Ocean tracer transport is represented by four processes: 1) eddy  
13 diffusion within the interior ocean ( $k$ ,  $3.2 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$ ); 2) deep upwelling in the interior  
14 ocean ( $w$ ,  $2.0 \times 10^{-8} \text{ m s}^{-1}$ ), which is balanced by lateral transport between the two surface  
15 layers as well as the down-welling in the polar deep ocean; 3) lateral exchange between  
16 the interior ocean and the well-mixed polar deep ocean ( $q$ ,  $7.5 \times 10^{-11} \text{ s}^{-1}$ ); and 4) vertical  
17 exchange between the high-latitude surface layer and the deep polar ocean ( $u$ ,  $1.9 \times 10^{-6} \text{ m}$   
18  $\text{s}^{-1}$ ) (Shaffer and Sarmiento 1995). The effective exchange velocity between surface  
19 ocean layers and the atmosphere in both low and high latitudes is assumed to be the same  
20 ( $2.32 \times 10^{-5} \text{ m s}^{-1}$ ) (Shaffer and Sarmiento 1995). Ocean carbonate chemistry (e.g., the  
21 Revelle buffer factor) is based on the formulation given by Sarmiento et al. (1992). In  
22 addition, we implemented the influence of sea surface temperature on the partial pressure  
23 of dissolved CO<sub>2</sub> in seawater with a sensitivity of  $\sim 4.3\% \text{ } ^\circ\text{C}^{-1}$  (Gordon and Jones 1973;



1 Takahashi et al. 1993; Joos et al. 2001). The changes in global mean sea-surface  
2 temperature (SST) is approximately 0.8-1.0 °C from 1850s to 2000s (Rayner et al. 2003;  
3 Brohan et al. 2006) slightly lower than that of the tropical land-based air temperature  
4 (~1.0 °C) but with a trend resembling the latter (Rayner et al. 2003; Jones et al. 2003;  
5 Hansen et al. 2006). For simplicity, therefore, we used the long-term trend of the tropical  
6 land air as a proxy for the corresponding trend in global SST.

7 The terrestrial biosphere in the Bern model is represented by four carbon compartments  
8 (ground vegetation, wood, detritus, and soil) with prescribed turnover rates and allocation  
9 ratios. The global net primary production (NPP), the influx to the biosphere, is assumed  
10 to be 60 GtC yr<sup>-1</sup> at the pre-industrial level; and the effect of CO<sub>2</sub> fertilization on NPP  
11 (i.e., the  $\beta$ -effect) is described with a logarithmic function with a  $\beta$  parameter of 0.38  
12 (Enting et al. 1994). The original Bern model does not consider the effects of changing  
13 global temperatures on terrestrial ecosystem respiration, which have been suggested to  
14 play an important role in regulating the variability of the global carbon cycle at  
15 interannual to multi-decadal time scales (Wang et al. 2013; Rafelski et al. 2009).  
16 Therefore, we implemented temperature's effects on terrestrial ecosystem respiration in  
17 the Bern model with an overall sensitivity ( $Q_{10}$ ) of ~1.5 (Lenton 2000; Davidson and  
18 Janssens 2006; Wang et al. 2013). We also changed the pre-industrial CO<sub>2</sub> concentration  
19 to 285 ppm in the Bern model to reflect the findings obtained from the observations (Fig.  
20 4 of the main text).

21 We calibrated the Bern model so that the model outputs fit the observed atmospheric CO<sub>2</sub>  
22 data most favorably. Because no major revisions were made to the ocean carbon cycle  
23 module (HILDA), we focused mainly on calibrating the biosphere module. With the

1 original biosphere model parameters, the simulated atmospheric CO<sub>2</sub> concentrations were  
2 found to be distinctly higher than observations, reaching ~411 ppm in 2010. These results  
3 are induced because rising temperatures enhance respiration in the model, reducing the  
4 net land carbon sinks to an unrealistic ~0.5 ppm yr<sup>-1</sup> in 2010. To balance the temperature-  
5 enhanced respiration, we need to increase the  $\beta$  parameter from 0.38 to 0.64 to  
6 incorporate a higher rate of gross biosphere carbon uptake as enhanced by CO<sub>2</sub>  
7 fertilization (Long et al. 2004) and the associated ecological changes (Keenan et al.  
8 2013). With the  $\beta$  parameter set at 0.64, the simulated global terrestrial NPP increased by  
9 14% from its pre-industrial level and reached ~69 GtC yr<sup>-1</sup> in 2010, which qualitatively  
10 agrees with recent estimates inferred from the isotope measurements (Welp et al. 2011).  
11 As such, the re-calibrated Bern model is able to simulate accurately the observed  
12 changes/variations in atmospheric CO<sub>2</sub> concentration and growth rate in the past 160  
13 years (Fig. 2 of the main text). The simulated ocean and land components of global  
14 carbon sinks are also consistent with estimates found in previous studies (e.g., Canadell et  
15 al. 2007; Le Quéré et al. 2009).

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## 17 A.2 Analysis of a General $N$ -Box Model

18 Eq. (1) in the main text can be generalized to describe an arbitrary  $N$ -component (“ $N$ -  
19 box”) carbon-cycle system:

$$20 \quad \dot{\mathbf{s}}' = \mathbf{X} \cdot \mathbf{s}' + \beta_T T' \cdot \mathbf{y} + \dot{E}' \cdot \mathbf{z}, \quad (\text{A1})$$

21 where  $\mathbf{s}'$ ,  $\mathbf{y}$ , and  $\mathbf{z}$  represent  $N \times 1$  vectors, and  $\mathbf{X}$  is an  $N \times N$  matrix. Specifically,

- 1 •  $s'$  represents all the anomalous carbon state variables (e.g., carbon in atmosphere,  
2 land, ocean, interior ocean, etc.). In particular, we assume the first element of  $s'$  to  
3 be the atmospheric carbon anomalies, that is,  $s'_1 = A'$  in Eq. (1) of the main text.
- 4 •  $y$  describes the distribution weights of the carbon impacts of temperature  
5 anomalies ( $\beta_T T'$ ) on different carbon pools. Per the reason explained in the main  
6 text, the elements of  $y$  are subject to the constraint  $\sum_{i=1}^N y_i = 0$ . Without the loss  
7 of generality we set  $y_1 = 1$ , reflecting that positive temperature anomalies release  
8 more carbon into the atmosphere.
- 9 •  $z$  describes the distribution weights of the  $\text{CO}_2$  anthropogenically emitted into the  
10 system. It is clear that  $z_1 = 1$  and  $z_i = 0$  ( $i = 2, \dots, N$ )
- 11 •  $X$  describes dynamics of and interactions among all the carbon reservoirs. In  
12 particular, the diagonal elements of  $X$  represent the decaying rates of the carbon  
13 reservoirs, i.e.,  $X_{ii} = -\alpha_i = -1/\tau_i$ . The off-diagonal elements  $X_{ij} \geq 0$  ( $i \neq j$ )  
14 represent the rates of carbon flow from the  $j$ -th reservoir to the  $i$ -th reservoir.

15  
16 *A2.1 Generalization of Eq. (5b) – the short-term responses of atmospheric CO<sub>2</sub> to an*  
17 *impulse disturbance of CO<sub>2</sub> emissions.*

18 It is easy to see that the characteristic equation of Eq. (A1) is given by

$$19 \quad \det(X - \lambda \cdot I) = 0, \quad (A2)$$

20 where  $\lambda$  is the vector of eigenvalues,  $I$  is the identity matrix, and “det” stands for the  
21 determinant of the matrix. By the binomial theorem, the sum of the eigenvalues equals  
22 the trace of the state matrix  $X$ , i.e.,

$$23 \quad \sum_{i=1}^N \lambda_i = \text{tr}(X) = \sum_{i=1}^N X_{ii} = \sum_{i=1}^N -\alpha_i, \quad (A3)$$

1 Because of the conservation of mass, the rank of  $X$  is  $N-1$  (see explanations in the main  
 2 text). Therefore, one of the eigenvalues is zero. We denote this zero-valued eigenvalue to  
 3 be  $\lambda_N$ . For simplicity of discussion, we also assume that the characteristic equation (A2)  
 4 does not have multiple roots (i.e., the state matrix  $X$  is not degenerated). It is clear that  
 5 this simplification has little influence on the discussion of the system's short-term  
 6 responses (i.e., when  $t \approx 0$ ).

7  
 8 The response function of atmospheric  $\text{CO}_2$  to a unit impulse of emission disturbances is  
 9 thus determined by

$$A'(t) = s'_1(t) = \sum_{i=1}^{N-1} \varphi_i \cdot \exp(\lambda_i t) + \omega_1, \quad (\text{A4})$$

10  
 11 where  $\omega_1$  is the steady-state (i.e., long-term) response of atmospheric  $\text{CO}_2$  to the  
 12 disturbance and  $\varphi_i$  are some constant coefficients subject to  $\sum_{i=1}^{N-1} \varphi_i + \omega_1 = 1$ , so that  
 13  $A'(0) = 1$ . Using the approximation that  $\exp(\lambda_i t) = 1 + \lambda_i t$  for  $\lambda_i t \approx 0$  in Eq. (A4) and  
 14 rearranging the items on the right-hand side we arrive at:

$$A'(t) = s'_1(t) \approx 1 + (\sum_{i=1}^{N-1} \varphi_i \lambda_i \cdot t) = \exp(-\alpha_{*,1} \cdot t), \quad (\text{A5})$$

15  
 16 where  $\alpha_{*,1} = \sum_{i=1}^{N-1} \varphi_i \lambda_i$ . Therefore,  $A'(t)$  initially decays as an exponential function.

17  
 18 We next prove that  $\alpha_{*,1}$  is indeed  $\alpha_1$ . By similar procedures as above, we can derive the  
 19 response functions of the  $i$ -th ( $i = 2, \dots, N$ ) carbon reservoir to be

$$s'_i(t) \approx \omega_i [1 - \exp(-\alpha_{*,i} \cdot t)], \quad (\text{A6})$$

20  
 21 where  $\omega_i$  is the steady-state (i.e., long-term) response of the specific reservoir. By mass  
 22 conservation it is apparent that  $\sum_{i=1}^N \omega_i = 1$ . For  $t \approx 0$ , all the responses of the surface

1 carbon reservoirs  $s'_i(t) \approx 0$  ( $i = 2, \dots, N$ ). Therefore the first equation in (A1) for the  
 2 atmospheric  $\text{CO}_2$  becomes

$$3 \quad \dot{A}' = \dot{s}'_1 \approx \mathbf{X}_{11} \cdot s'_1 = -\alpha_1 A', \quad (\text{A7a})$$

4 which simply means that

$$5 \quad \alpha_{*,1} = \alpha_1, \text{ and } A'(t) \approx \exp(-\alpha_1 \cdot t), \quad (\text{A7b})$$

6 which is the same conclusion stated by Eq. (5b) in the main text (where  $\alpha_1$  is denoted by  
 7  $\alpha_A$ ).

8  
 9 Finally, because the trace of the state matrix  $\mathbf{X}$  is invariant under unitary transforms,  
 10 rearranging/re-combining the surface carbon reservoirs will not change the results of Eq.  
 11 A8. This further proves the generality of Eq. (5b) in the main text.

12  
 13 *A2.2 Generalization of Eq. (3b) – the long-term responses of atmospheric  $\text{CO}_2$  to an*  
 14 *impulse disturbance of  $\text{CO}_2$  emissions.*

15  
 16 At the steady state Eq. (A1) becomes

$$17 \quad \mathbf{X} \cdot \mathbf{s}' = 0, \quad (\text{A8})$$

18 with  $\sum_{i=1}^N s'_i = 1$  (mass conservation). This condition indicates that Eq. (A8) has a non-  
 19 trivial solution  $\mathbf{s}'^*$ , which can be found by well-known procedures of linear algebra. With  
 20 the solution  $\mathbf{s}'^*$ , by the first row of Eq. (A8) it is clear that

$$21 \quad A'/\tau_A = \mathbf{X}_{11} \mathbf{s}'^*_1 = \sum_{i=2}^N \mathbf{X}_{1i} \mathbf{s}'^*_i = S'/\tau_S, \quad (\text{A9})$$

22 where  $S' = \sum_{i=2}^N \mathbf{s}'^*_i$ , denoting the total responses of all the surface carbon reservoirs;  
 23 and  $\tau_S = S'/\sum_{i=2}^N \mathbf{X}_{1i} \mathbf{s}'^*_i$ , representing the “bulk” response time constant of the surface

1 carbon reservoirs. Eq. (A10) is the same as Eq. (3b) in the main text, that is, the  
 2 relationship is accurate for general carbon dynamic system if we have complete  
 3 knowledge of  $\mathbf{X}$ .

4  
 5 Because in reality we do not have accurate information of  $\mathbf{X}$ , the estimates of  $\tau_S$  (and  $\tau_A$ )  
 6 are associated with uncertainties. To illustrate the difficulty of the problem, we consider a  
 7 particular N-box system that includes only the atmosphere and the global oceans at  
 8 different layers. Let  $s'_1 = A'$  and  $s'_i$  ( $i = 2, \dots, N$ ) denote different ocean layers with  
 9 increasing “ $i$ ” indicating increasing ocean depth. We further assume that each ocean layer  
 10 only interacts with its neighbors and the atmosphere only interacts with the surface ocean  
 11 layer. As such, the state matrix  $\mathbf{X}$  can be written as:

$$12 \quad \mathbf{X} = \begin{bmatrix} -\alpha_1 & \beta_2\alpha_2 & 0 & 0 & \cdots & 0 \\ \alpha_1 & -\alpha_2 & \beta_3\alpha_3 & 0 & \cdots & 0 \\ 0 & (1-\beta_2)\alpha_2 & -\alpha_3 & \beta_4\alpha_4 & \cdots & \vdots \\ 0 & 0 & (1-\beta_3)\alpha_3 & \ddots & \ddots & 0 \\ \vdots & \vdots & \vdots & \ddots & -\alpha_{N-1} & \alpha_N \\ 0 & 0 & \cdots & 0 & (1-\beta_{N-1})\alpha_{N-1} & -\alpha_N \end{bmatrix} \quad (A10)$$

13 where  $\alpha_i = 1/\tau_i$  and  $\beta_i$ s are constant numbers in the range  $[0, 1]$ . Thus  $\beta_i$  and  $(1-\beta_i)$   
 14 reflect the relative weights of the carbon efflux of  $i$ -th reservoir to its two neighbors. Note  
 15 that  $\beta_1 = 0$  and  $\beta_N = 1$ .

16  
 17 Solving the steady-state equation (A8) with the  $\mathbf{X}$  of Eq. (A10), we obtain the results

$$18 \quad s'^*_i = \prod_{j=2}^i \frac{(1-\beta_{j-1})}{\beta_j} \cdot \frac{\alpha_1 A'}{\alpha_i} = \prod_{j=2}^i \frac{(1-\beta_{j-1})}{\beta_j} \cdot \frac{\tau_i A'}{\tau_1} \quad (A11a)$$

19 and by Eq. (A9), we estimate  $\tau_S$  to be

$$20 \quad \tau_S = \sum_{i=2}^N \left( \prod_{j=2}^i \frac{(1-\beta_{j-1})}{\beta_j} \cdot \tau_i \right) \quad (A11b)$$

1 Therefore,  $\tau_S$  is not only a function of  $\tau_i$  but also of  $\beta_j$ . In particular, the (relative)  
 2 sensitivity of  $\tau_S$  to  $\beta_2$  is

$$3 \quad (d\tau_S/d\beta_2)/\tau_S = -1/[\beta_2(1 - \beta_2)]. \quad (A12)$$

4 Because of the characteristic buffering effect of the ocean carbonate chemistry, the  
 5 anomalous carbon exchange (induced by anthropogenic disturbances) between ocean  
 6 surface and the atmosphere is much (~10 times) more effective than between ocean  
 7 surface and deep oceans (Gruber and Sarmiento 2002). This means that the value of  $\beta_2$  is  
 8 close to 0.9 or  $(d\tau_S/d\beta_2)/\tau_S \approx 10$ . Therefore, a 1% uncertainty in  $\beta_2$  alone could induce  
 9 10% uncertainty in  $\tau_S$  (or  $S'$ )!

10  
 11 The above example highlights the challenge in estimating  $\tau_S$  and thus the long-term  
 12 response of the atmospheric  $CO_2$  to anthropogenic emission disturbances. This problem is  
 13 particularly emphasized in the main text (Line 10-26, Page 13968). We argued that the  
 14 problem is mainly induced by the limited observations of the global climate-carbon  
 15 system such that our knowledge of the state matrix  $\mathbf{X}$  is incomplete.

16  
 17 A2.3 Generalization of Eq. (6b) – the long-term responses of atmospheric  $CO_2$  to a unit  
 18 step change of global surface temperature

19  
 20 It is clear that the steady state Eq. (A1) for temperature disturbance is

$$21 \quad -\mathbf{X} \cdot \mathbf{s}' = \beta_T T' \cdot \mathbf{y}. \quad (A13a)$$

22 By the notions developed in Eq. (A9), we can represent the first row of (A13a) as

$$23 \quad \alpha_A A' - \alpha_S S' = \beta_T T', \quad (A13b)$$

1 where  $\alpha_A = 1/\tau_A$  and  $\alpha_S = 1/\tau_S$ .

2

3 Because changes in temperature do not induce changes in the total carbon of the system,

4 it means that

5 
$$A' + S' = 0, \text{ or, } A' = -S', \text{ \underline{\hspace{10em}} (A14)}$$

6 Substituting Eq. (A14) in Eq. (A13b) and rearranging the items, we obtain

7 
$$A' = \beta_T / (\alpha_A + \alpha_S) \cdot T' \approx \beta_T / \alpha_A \cdot T', \text{ \underline{\hspace{10em}} (A15)}$$

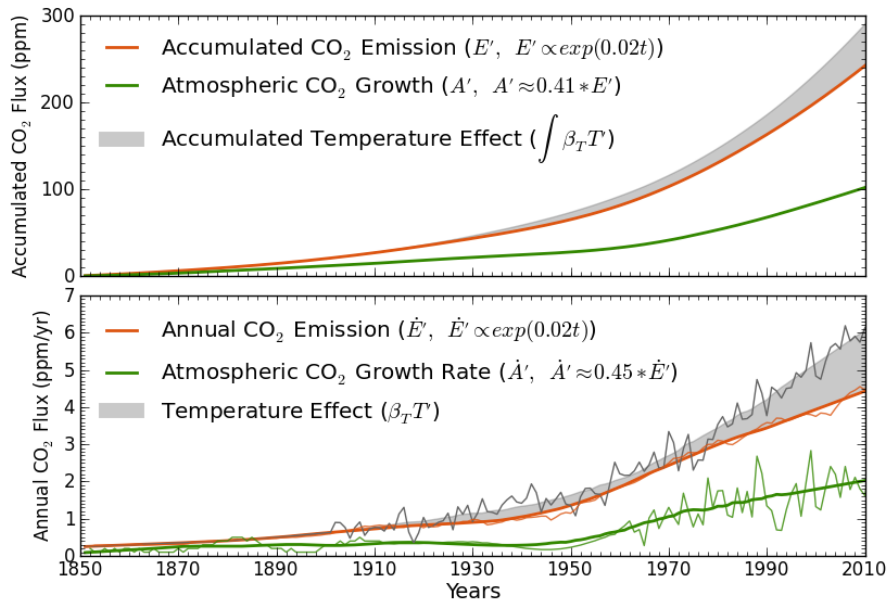
8 for generally  $\alpha_A \gg \alpha_S$ . This is the same equation as Eq. (6b) in the main text.

9

10



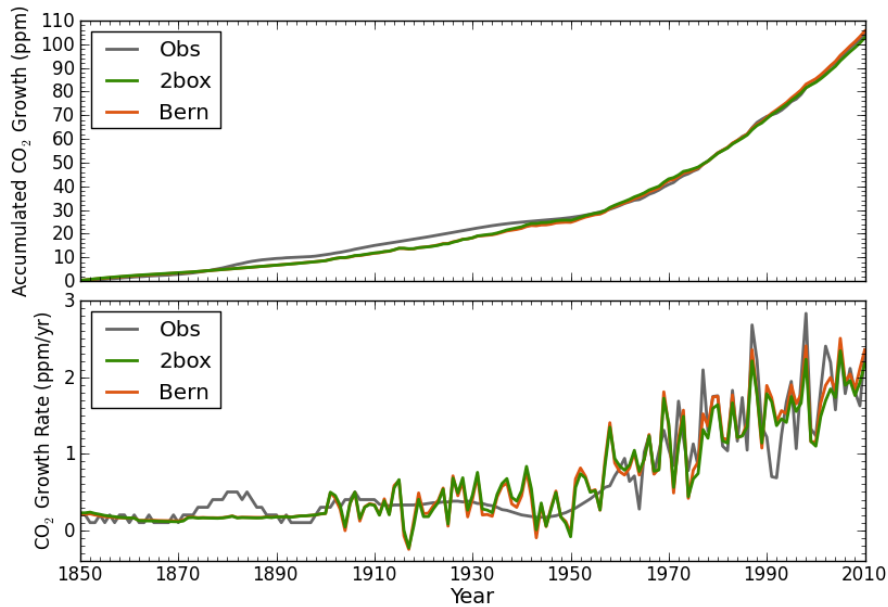
1 **Figures**



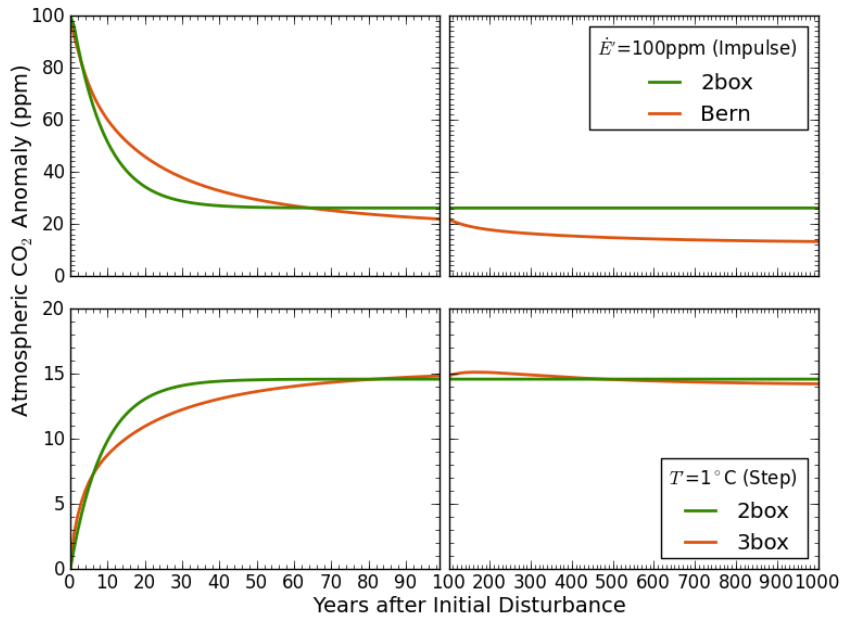
2  
 3 Fig. 1 Time series of global anthropogenic CO<sub>2</sub> emissions (red line), atmospheric CO<sub>2</sub>  
 4 concentrations (green line), and the anomalous CO<sub>2</sub> fluxes induced by warming surface  
 5 temperatures (gray shade) between 1850 and 2010. The Top panel indicates the  
 6 accumulated CO<sub>2</sub> fluxes or the total concentration changes while the Bottom panel shows  
 7 them at annual steps. The thick and the thin lines indicate long-term and interannual  
 8 variations of the time series, respectively. The mathematical symbols are the same as in  
 9 Eq. (1) and explained in the text. In both annual and accumulative cases, CO<sub>2</sub> emissions  
 10 largely increase as an exponential function of time, while changes in the atmospheric CO<sub>2</sub>  
 11 concentrations are proportional to the corresponding emissions by a factor about 0.41-  
 12 0.45.

13

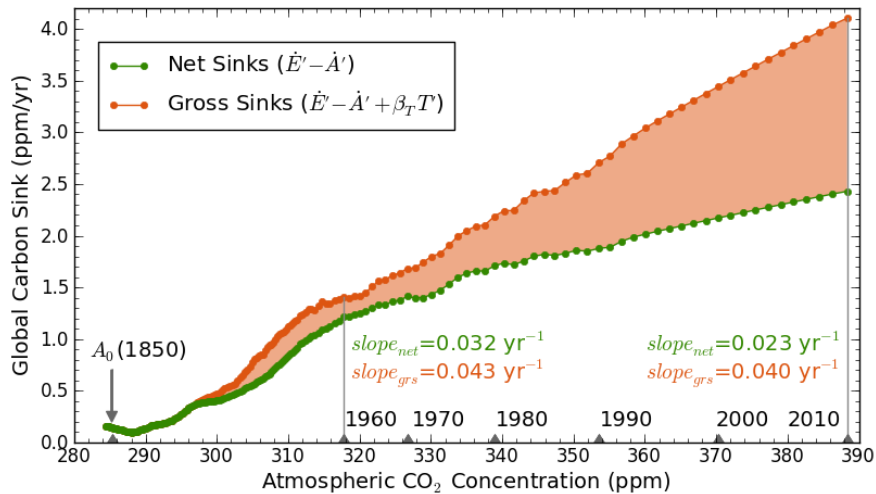
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1  
2 Fig. 2 Simulations of the observed atmospheric CO<sub>2</sub> concentrations (Top Panel) and  
3 growth rates (Bottom Panel) from anthropogenic CO<sub>2</sub> emissions and land-surface air-  
4 temperature data using the two-box model (“2box”) and the [revised](#) Bern model (“Bern”).  
5 The atmospheric CO<sub>2</sub> concentration in 1850 (i.e., 284.7 ppm) is used as the initial  
6 condition for the model integration. Long-term mean temperature before 1901 is assumed  
7 to be stable and represented by the 1901-1920 mean. Other model parameters used in  
8 these simulations are explained in the main text (the two-box model) or the Appendix  
9 (the [revised](#) Bern model).  
10



1  
 2 Fig. 3 Disturbance-response functions of the atmospheric CO<sub>2</sub> concentration simulated by  
 3 the two-box model (“2box”) and the revised Bern model (“Bern”). The Top panel shows  
 4 the responses of atmospheric CO<sub>2</sub> concentration to an impulse increase (of 100 ppm) in  
 5 anthropogenic CO<sub>2</sub> emissions and the Bottom panel shows the corresponding responses  
 6 to a step increase (of 1 °C) in surface temperatures.  
 7



1  
 2 Fig. 4. Global annual carbon sinks (ppm/yr) as a function of atmospheric CO<sub>2</sub>  
 3 concentration from 1850 to 2010. The green dots indicate the observed “net” carbon sinks  
 4 and the red dots indicate the “gross” carbon sinks that accounted for the effects of  
 5 temperature changes (Eq. 7). The differences between the gross and the net carbon sinks  
 6 (the shaded area) indicate the extra carbon fluxes released into the atmosphere as a result  
 7 of warming temperatures (Fig. 1). The gray arrow (“A<sub>0</sub>”) indicates the estimated  
 8 atmospheric CO<sub>2</sub> level (284.7 ppm) that was stable at pre-industrial CO<sub>2</sub> emission rates  
 9 and climate conditions. The slopes between the global annual carbon sinks and  
 10 corresponding changes in atmospheric CO<sub>2</sub> concentration (relative to A<sub>0</sub>) generally reflect  
 11 carbon-sequestration efficiencies of global land and ocean reservoirs.  
 12

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