Modeling of Atmospheric CO$_2$ Concentrations as a Function of Carbon Dioxide Emissions with Implications for the Fossil-Fuel Atmospheric Fraction (AF$_{FF}$)

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Abstract: In this work, a semi-empirical relationship of carbon dioxide emissions with atmospheric CO$_2$ concentrations has been developed that is capable of closely replicating observations from 1751 to 2018. The correlation consists of a superposition of a linear component that may be attributed to the net emission flux from land use changes coupled with a rapidly varying component of the terrestrial sink combined with a fossil-fuel combustion/cement production emissions-based calculation with a single, fixed, scaling parameter determined by the ocean sink coupled with the remaining slowly varying component of the land sink (the fossil-fuel combustion airborne fraction).

Keywords
Carbon dioxide emissions; Carbon dioxide concentrations; Atmospheric Fraction

1. Background & Historical Data

In 1861, Irish physicist John Tyndall presented results from his measurements on the absorption of “calorific rays” by various gases to the Royal Society (Fleming 1998; Tyndall 1861). This presentation is believed to be the first attribution of atmospheric gases, and specifically of water vapor and carbon dioxide, to changes in the climate. Since that time and, most especially, for the past several decades, there has been a significant focus upon the emissions of carbon dioxide into the atmosphere and the potential impact of increasing atmospheric carbon dioxide concentrations upon the global climate (see, e.g., Oreskes 2004; Prentice 2001; Meehl 2007). In this work, a simple semi-empirical parameterization is presented that precisely reproduces the increase of atmospheric CO$_2$ concentrations observed from 1751 to 2018 resulting from emissions of carbon dioxide into the environment. The implications of this analysis for the determination of the atmospheric fraction due to fossil fuel consumption (AF$_{FF}$) are discussed.

Sources of anthropogenic carbon emissions from fossil fuel combustion, including gas flaring and cement production are collected and reported by a number of organizations (Andres 2012) including the Carbon Dioxide Information Analysis Center [CDIAC], the International Energy Agency [IEA], the United Nations [UN], and the United States Department of Energy [DoE] Energy Information Administration [EIA]. Similar to Le Quéré et al. (Le Quéré 2018), the current investigation utilizes the CDIAC data set (Boden 2013, Boden 2018) of energy-based, human-caused carbon emissions since 1751. The data include total carbon emissions from fossil fuel consumption and cement production and are shown in Fig. 1(a). As may be observed, emissions have been increasing steadily for over 250 years and more particularly, there has been a substantial increase in emissions since 1950 that has continued to the present.
Figure 1. (a) CO$_2$ Emissions into the Environment from Fossil Fuel Consumption and Cement Production (Boden 2013, Boden 2018) (Note: logarithmic scale on y-axis).

(b) Observations of atmospheric CO$_2$ concentrations (ppm) (Etheridge 1998; Neftel 1994; Tans 2018; Keeling 2018).

Fig. 1(b) displays measurements of the CO$_2$ concentration in the atmosphere based on the Law Dome (Etheridge 1998) and Siple (Neftel 1994) ice cores and direct measurements at Mauna Loa (Tans 2018; Keeling 2018). These data demonstrate that the carbon dioxide concentration has been increasing steadily since 1750 and this increase has also accelerated significantly since 1960.

Efforts to associate the change in concentration with emissions based on a variety of models have been attempted. Many of these studies examined the uptake of carbon dioxide by the ocean as it is a large carbon sink in the environment. Examples include; study of the magnitude, variability and trends in the global ocean carbon uptake (Wanninkhof 2013), examination of feedback mechanisms and sensitivities of ocean carbon uptake under global warming (Plattner 2001), and reconstructions of the history of anthropogenic CO$_2$ concentrations in oceans (Khatiwala 2009). These studies demonstrate that the CO$_2$ uptake into the ocean has been increasing over the past several decades.
and, at present, approximately 1 gram of CO\textsubscript{2} is absorbed by the ocean for every 4 grams emitted into the environment (Le Quéré 2018).

Correlation of Atmospheric CO\textsubscript{2} Concentration with Carbon Dioxide Emissions

2. Traditional Approach & Discussion

The growth of carbon emissions from land use changes coupled with the burning of fossil fuels is shown in Figure 2 [Boden 2013, Boden 2018, Stocker 2014, Le Quéré 2018]. It is noted that an exponential growth curve will generally follow the overall shape of the observed data set, notwithstanding significant differences of up to 38% from the observations. It has been postulated that if the climate system is considered as a linear system forced by exponentially growing carbon dioxide emissions, then all ratios of responses to forcings are constant [Raupach 2013]. In particular, the atmospheric fraction, AF, would be constant with the value dependent upon the lifetime, \( \tau \), of the CO\textsubscript{2} in the atmosphere [Terenzi and Khatiwala, 2009]. A best fit to the exponential curve yields a value of approximately 43% for AF [Terenzi and Khatiwala, 2009]. AF has been extensively discussed in the literature (see, e.g., Jones 2005, Canadell 2007, Raupach 2008, Knorr 2009). These works indicate AF values of approximately 40±14% (Jones 2005) with a possible slight upward trend noted per decade (Canadell 2007, Raupach 2008). However, this upward trend was not reported Knorr (2009).

![Figure 2. Levels of CO\textsubscript{2} from Fossil Fuels and Land-Use Changes.](image)

To convert the emissions data from Fig. 2 to changes in atmospheric CO\textsubscript{2} concentrations, a measured base-year [2018] concentration datum of 408.52 ppm was chosen from the Mauna Loa carbon dioxide measurements data set (Tans 2018; Keeling 2018). The change in the atmospheric CO\textsubscript{2} concentration was then determined by using the combined fossil-fuel based (Boden 2013, Boden 2018) and land-use changes induced CO\textsubscript{2} emissions (Stocker 2013, Le Quéré 2018) for each year preceding 2018, converting those annual emission rates into an equivalent ppm of the atmosphere [mass of the atmosphere=5.148E18 kg (Trenberth 2005)], and then applying a single scaling factor (airborne...
fraction (AF) for each year [43.1%] to determine the concentration change for that year. For each year prior to 2018, the change was negative. The results are shown in Fig. 3.

![Predicted vs Observed CO₂ Atmospheric Concentrations AF = 43.1%](image)

**Figure 3.** Predicted CO₂ Concentrations with Observations using AF=43.1%.

The curve in Fig. 3 depicting the predicted concentrations follows the CO₂ concentration observations reasonably well but is not a precise match. To examine this further, it is illustrative to examine the carbon dioxide emissions more closely as a function of time to determine if an exponential growth curve assumption for emissions is warranted and hence, implying a constant value for AF. Figure 4 shows the land use carbon dioxide emissions (Stocker 2013, Le Quéré 2018). A cursory examination of these data clearly demonstrates the emissions do not follow an exponential growth pattern.

![Land Use Changes CO₂ Emissions](image)

**Figure 4.** Emissions from Land-Use Changes.

Now, consider fossil-fuel based emissions in the time period from 1751 to 2018 (Boden 2013, Boden 2018), shown in Figure 5. As may be seen, the agreement between the best fit exponential
growth curve with the data are poor in the early 20th century and significantly worse post-1980. Although the observations from the nineteenth century (Fig. 5(b)) clearly follow an exponential growth curve (note the logarithmic scale in Fig. 5(b)), the data post-1945 are best fit by a linear growth curve (Fig. 5(c)).

Figure 5. Comparison of Best Fit Exponential Growth Curve with Fossil Fuel and Cement Production CO₂ Emissions Observations.

(a) Comparison of Best Fit Exponential Growth Curve with Fossil Fuel and Cement Production CO₂ Emissions Observations
Comparison of Best Fit Linear Growth Curve with Fossil Fuel and Cement Production CO\textsubscript{2} Emissions Observations.

Although there is moderate agreement in Fig. 3 between the CO\textsubscript{2} concentration observations with those predicted with a fixed atmospheric fraction, AF, of 43\%, the deviations from exponential growth of both the carbon emissions from land use changes (Fig. 4) and from fossil fuels (Fig. 5) suggest that the approximate concurrence observed in Fig. 2 of the sum of the emission with an exponential growth curve is coincidental and not indicative of a fundamental physical nature of the emissions. This issue raises concerns with the postulate [Raupach 2013] that if the climate system is considered as a linear system forced by exponentially growing carbon dioxide emissions, then all the ratios of responses to forcings, including the atmospheric fraction, are constant.

It is also illustrative to consider that from 1750 to 1850, the total carbon dioxide emissions (FF + LUC) equaled 71.7 Gt CO\textsubscript{2} with 93.4\% of those emissions due to land use changes (Stocker 2013, Boden 2013, Boden 2018). Using the approach described above to convert emissions to CO\textsubscript{2} concentration changes, this equates to a 9.2 ppm increase of CO\textsubscript{2} in the atmosphere if all the emitted CO\textsubscript{2} remained in the atmosphere. The measured data (Etheridge 1998, Neftel 1994) indicate a 9.8 ppm increase in carbon dioxide atmospheric concentrations during that period. So, for 100 years at the beginning of the industrial era, this concurrence implies that ~100\% of the total CO\textsubscript{2} emissions remained in the atmosphere. This somewhat surprising result raises the question: why is there apparently no evidence of an ocean or land-based carbon sink during this century-long time span?

3. Alternative Correlation Approach

In the same manner as used to generate Fig.3, a measured base-year [2018] concentration datum of 408.52 ppm was chosen from the Mauna Loa carbon dioxide measurements data set (Tans 2018; Keeling 2018). The change in the atmospheric CO\textsubscript{2} concentration was then determined by using only the fossil-fuel based CO\textsubscript{2} emissions (Boden 2013, Boden 2018) for each year preceding 2018, converting those annual emission rates into an equivalent ppm of the atmosphere, and then applying a single scaling factor (airborne fraction due to fossil fuel consumption, (AF\text{ff})) for each year [best fit: 54.5\%] to determine the concentration change for that year. For each year prior to 2018, the change was negative. The results are shown in Fig. 6.
Figure 6. Measured and Predicted Atmospheric Carbon Dioxide Concentrations
Predicted Values Use 2018 Datum as Base Year; Scaling Factor of 54.5% (AFFF) applied to Emissions Calculation for each year.

As may be observed, there is excellent agreement between the predicted values and experimental observations from approximately 1925 to 2018. The agreement before 1925 is less precise because the predicted values become asymptotic as the carbon dioxide emissions from fossil fuel combustion and cement production diminish significantly during the nineteenth and eighteenth centuries while the measured atmospheric CO₂ concentration values from the Siple (Neftel 1994) and Law Dome (Etheridge 1998) ice cores continue a steady decline as one moves backward in time from about 1925 to 1750.

Figure 7. CO₂ Concentrations with No Fossil fuel combustion/cement production emissions inputs
(Note: 1839 data point removed from analysis).
To further refine the correlation of emissions with concentration, a closer examination of the measured CO$_2$ concentration data from 1750 to 1898 was performed. As may be seen in Fig. 7, the increase in the non-fossil fuel/cement production-driven carbon dioxide atmospheric concentration over the 148-year period is well characterized by a simple linear function. One may now characterize the overall change in atmospheric CO$_2$ concentration since 1750 as a superposition of the linear increase shown in Fig. 7 extended through the present combined with the delta induced by fossil fuel/cement production-driven carbon dioxide emissions using an approach similar to that employed in Fig. 6. When implemented, the sole empirical scaling factor, the airborne fraction due to fossil fuels (AF$_{FF}$), shifts to 51.3% from the 54.5% factor deduced for the results shown in Fig. 6. The resultant agreement to the measured data is shown in Fig. 8.

![Figure 7](image)

**Figure 7.** Predicted and Observed Atmospheric CO$_2$ Concentrations

An analysis of the statistical validity of the fit of the semi-empirical model to the measured data was performed using Anova in Microsoft Excel (Microsoft 2018). A linear regression of the predicted atmospheric CO$_2$ values versus the measured CO$_2$ levels yields the results shown in Figure 9. The slope of 1.008 (R-square: 0.9995) demonstrates almost a perfect fit (exact would be a slope of 1.000) with a robust statistically significant relationship (p-value: 3.2E-140). This result provides strong statistical evidence that the airborne fraction of fossil-fuel-based CO$_2$ emissions, AF$_{FF}$, has been unchanged at 51.3% for the entire analysis period of 268 years.
Discussion

Using the terminology of Le Quéré et al. (Le Quéré 2018), the global carbon budget is a balance of emission and absorption processes. This balance equation may be written as:

\[ E_{FF} + E_{LUC} = G_{ATM} + S_{OCEAN} + S_{LAND} + B_{IM} \]

where \( E_{FF} \) is the estimate for CO\(_2\) emissions from fossil fuel combustion and cement production; \( E_{LUC} \) is the estimate for CO\(_2\) emissions resulting from deliberate human activities on land; \( G_{ATM} \) is the growth rate of CO\(_2\) in the atmosphere; \( S_{OCEAN} \) is the uptake of CO\(_2\) in the ocean; \( S_{LAND} \) is the uptake of CO\(_2\) by the terrestrial sink; and, \( B_{IM} \) is an estimate of the budget imbalance, which is a measure of the mismatch between the estimated emissions and the estimated changes in the atmosphere, land, and ocean.

Referring to Fig. 1(a), one observes significant increases in fossil-fuel combustion and cement production carbon dioxide emissions, \( E_{FF} \), since the mid-1700s and since 1959, emissions have increased 370% (Boden 2013, Boden 2018). Models of the ocean sink, \( S_{OCEAN} \), have also shown increases from 5.5±1.8 GtCO\(_2\) during the decade of the 1960s to 9.2±1.8 GtCO\(_2\) from 2000 to 2009 (Le Quéré 2018; Wanninkhof 2013; Plattner 2001; Khatiwala 2009). These increases have smoothly varied over time (see Fig. 3 of (Le Quéré 2018)). In contrast, while the land sink, \( S_{LAND} \), has also increased, there has been dramatic variability in CO\(_2\) absorption over short time periods estimated using either Dynamic Global Vegetation Models (DGVM) (Lawrence 2011; Levy 2004; Clark 2011; Cox 2001; Sitch 2003; Smith 2001; Ahlström 2012; Zaehle 2011; Krinner 2005; Woodward 2004; Zeng 2005) or using the residual from Equation 1 with inputs of measured & modeled \( E_{FF} \), \( E_{LUC} \), \( G_{ATM} \) and \( S_{OCEAN} \).

To account for the significant interannual variability in the land sink, it may be hypothesized that the terrestrial carbon sink is a combination of two elements; one component that is slowly varying that responds to smooth changes in the emissions of carbon dioxide coupled with a reactive component that responds to rapid changes in emissions and is likely correlated with the rapid changes in vegetation considered in the DGVMs. This approach would enable the terrestrial sink to adapt to rapid changes in land-based emissions (and correlate with the significant variation in interannual terrestrial sink observations) while also accounting for a fraction of the more smoothly varying fossil-fuel based emissions.

As shown in Fig. 8, it is possible to fully characterize the change in carbon dioxide concentrations over a 268-year period using only one measured concentration datum [2018-Mauna Loa (Tans 2018; Keeling 2018)] combined with a linear regression fit to non-fossil fuel/cement production-driven
induced CO₂ concentration increases and a fixed-parameter scaled, AF₉₅, calculation of the changes in CO₂ concentration due to fossil fuel combustion and cement production emissions prior to 2018. Based on these data, it is suggested that Eq. 1 may be used to determine the net flux of land-based CO₂ emissions, E₉₅, combined with the reactive component of the terrestrial sink, S₉₅ from 1750 AD to the present. As observed in Fig. 7, G₉₅ increased linearly at a modest rate during the selected time period; approximately 0.099 ppm per year. Converting this increase to a net emissions rate, it is determined that the net flux of E₉₅ minus the rapidly-varying component of S₉₅ is 0.77 GtCO₂ per year. The remainder of the land sink coupled with the ocean sink operates as a smoothly varying function that absorbs 48.7% of the emissions from fossil fuels and cement production. This somewhat non-intuitive hypothesis is borne out by the data.

The present work has determined to a strong statistical validity that the airborne fraction due to fossil fuel consumption (AF₉₅) has been unvarying at 51.3% for the past 268 years. Since neither fossil-fuel nor land use change based carbon emissions follow an exponential growth curve for that time period, postulates based on the assumption of exponential emission growth (Raupach 2009) may not be valid. Thus, the variations in the inputs to the global carbon budget suggest that the determination of a single sink scaling factor of 48.7% for fossil-fuel energy-based CO₂ emissions over such an extended period may be considered significant. If one studies the increases and decreases over time in the broad range of constituents influencing the climate (see, e.g., Fig. 1.1 from the IPCC AR5 report (Stocker 2013)) [coupled with the non-exponential growth patterns of land-based or fossil-fuel driven emissions negating the eigenvector approach of Raupach (2008)], it may not be expected that a constant AF₉₅ applied to the measured fossil-fuel-based carbon dioxide emissions (Boden 2013, Boden 2018) would accurately reproduce the measured changes in carbon dioxide concentrations.

5. Summary

A semi-empirical correlation between carbon dioxide emissions with CO₂ concentrations has been developed that is capable of closely replicating observations from 1751 to 2018. The key characteristics of the correlation are a superposition of a linear variation that may be attributed to the net flux of land use changes with the rapidly-varying reactive component of the terrestrial sink coupled with a fossil-fuel combustion/cement production emissions-based calculation with a single, fixed scaling parameter (AF₉₅) driven by the ocean sink and the smoothly varying component of the terrestrial sink. Additional research is necessary to determine how the wide array of parameters inputting into carbon dioxide concentrations results in a value for AF₉₅ that has been unvarying over the past 268 years.

**Author Contributions:** This work is the sole effort of John P. O’Connor.

**Competing Financial Interests:** The author reports no competing financial interests.

**Data Availability Statement:** The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.
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