

SOURCES AND SINKS OF CARBON DIOXIDE

Tom Quirk

twquirk@labyrinth.net.au

ABSTRACT

The conventional representation of the impact on the atmosphere of the use of fossil fuels is to state that the annual increases in concentration of CO₂ come from fossil fuels and the balance of some 50% of fossil fuel CO₂ is absorbed in the oceans or on land by physical and chemical processes.

An examination of the data from:

- i) measurements of the fractionation of CO₂ by way of Carbon-12 and Carbon-13 isotopes,*
- ii) the seasonal variations of the concentration of CO₂ in the Northern Hemisphere and*
- iii) the time delay between Northern and Southern Hemisphere variations in CO₂,*

raises questions about the conventional explanation of the source of increased atmospheric CO₂.

The results suggest that El Nino and the Southern Oscillation events produce major changes in the carbon isotope ratio in the atmosphere. This does not favour the continuous increase of CO₂ from the use of fossil fuels as the source of isotope ratio changes. The constancy of seasonal variations in CO₂ and the lack of time delays between the hemispheres suggest that fossil fuel derived CO₂ is almost totally absorbed locally in the year it is emitted. This implies that natural variability of the climate is the prime cause of increasing CO₂, not the emissions of CO₂ from the use of fossil fuels.

1 INTRODUCTION

The conventional representation of the impact on the atmosphere of the use of fossil fuels is to state that the annual increases in concentration of atmospheric CO₂ come from fossil fuel emissions and the balance of some 50% of emissions is absorbed in the oceans or on land by chemical or biogenic interactions¹. The annual changes at Mauna Loa² are shown in Figures 1A and 1B along with the estimated production of CO₂ from fossil fuels³. Note that the absorption shown in Figure 1B is simply found by subtraction of the atmospheric increase from the calculated emissions of CO₂ shown in Figure 1A.

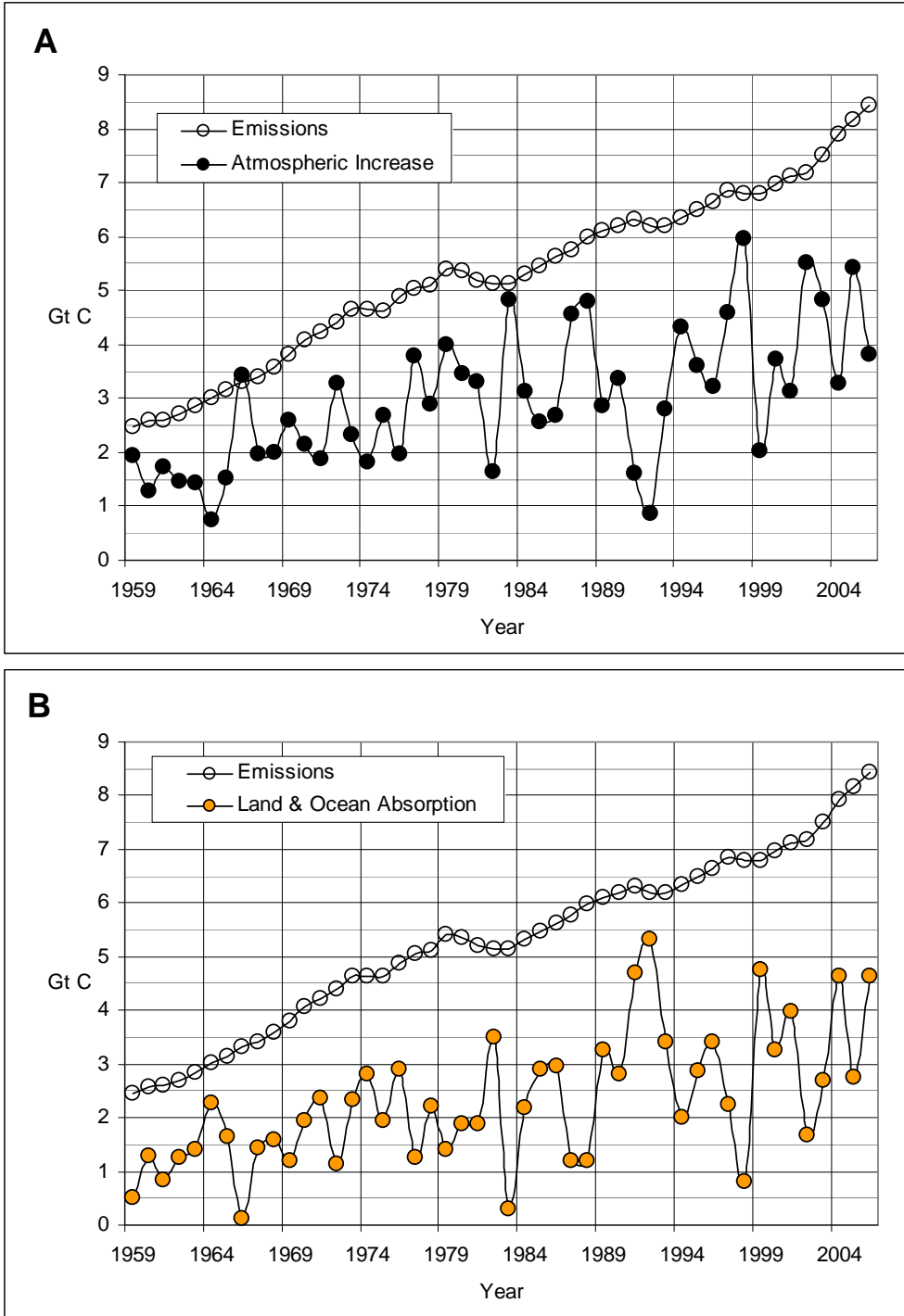


Figure 1 A) Estimated emissions from worldwide fossil fuel use and the annual increase in atmospheric CO₂ sourced from <http://cdiac.ornl.gov/trends/co2/contents.htm>. **B)** Estimated emissions from worldwide fossil fuel use and the estimated amount of CO₂ absorbed by land and ocean, calculated by subtraction of the atmospheric CO₂ increase from the estimated emissions from fossil fuels.

The justification for this conclusion is supported by measurements of the falling proportion of ^{13}C in the atmosphere which is taken to signal the appearance of CO_2 from fossil fuel emissions.

The fundamental assumption of the anthropogenic source of increasing CO_2 is the basis for all climate change policy development. It has deep historical roots given fresh force from measurements started in the late 1950's. Extended time series data have become available and it is well worth reviewing the data to check that original assumptions remain valid.

Carbon has two stable isotopes, Carbon-12 (^{12}C) and Carbon-13 (^{13}C). In general ^{12}C is 99% of natural carbon and ^{13}C 1%. However the rates at which chemical reactions and physical processes take place vary due to the masses of the isotopes present. For carbon the result is fractionation of the isotopes so that carbon found in coal or oil is 1.083% ^{13}C while carbon in the atmosphere is 1.103% ^{13}C .

Results are given as $\delta^{13}\text{C}$, a measure of the isotope ratio of $^{13}\text{C}/^{12}\text{C}$ defined as:

$$\delta = 1000 \times ((^{13}\text{C}/^{12}\text{C})_{\text{sample}} / (^{13}\text{C}/^{12}\text{C})_{\text{standard}} - 1)$$

The standard ratio is an international standard of comparison and is the zero point for δ ⁴.

δ for CO_2 coming from biogenic sources has an approximate value of -26⁵, the fossil fuel value is -29, (estimated from US energy use⁶ from a range of -20 to -44) and CO_2 from the ocean is given values ranging from -7 to 1.5⁷. The atmospheric values are found to vary from -7 to -8. If there are two sources of CO_2 , the ocean and a biogenic or fossil fuel source then the average implies that some 4% to 33% of atmospheric CO_2 has been derived from biogenic or fossil fuel sources⁸.

One of the great unknowns is the variability of the oceans that have multi-year cycles that cause temperature variations such as El Nino and the Southern Oscillation events. It is not clear whether there are other climate effects from the oceans and from the land and ocean based sources of biogenic CO_2 . However the size of these reservoirs and the annual exchanges with the atmosphere are estimated to be at least a factor of 20 times the annual production of CO_2 from fossil fuel⁹. There are substantial uncertainties in reservoir size and exchanges.

This paper provides an analysis of the changes in CO_2 and ^{13}C in the atmosphere using the extensive measurements made by the SIO¹⁰ and the CSIRO¹¹ spanning many years. All the data are available on the website <http://cdiac.ornl.gov/trends/co2/contents.htm> with CO_2 concentrations and $^{13}\text{C}/^{12}\text{C}$ isotope ratios in monthly and yearly form^{12,14}. The development of this extensive set of measurements has been reviewed recently by Beck¹³.

Three approaches have been adopted, first looking directly at the variations of ^{13}C in the atmosphere, second, looking at the seasonal variations of CO_2 through the annual summer-winter cycles and finally examining timing differences of the Northern and Southern Hemisphere changes in CO_2 .

2 CARBON-13 IN THE ATMOSPHERE

Variations in the amount of ^{13}C in CO_2 are shown in Figures 2 and 3 with data from the Scripps Institute of Oceanography (SIO) for Mauna Loa and also the South Pole¹⁴.

Variations in ^{13}C have a strong cyclic pattern in the Northern Hemisphere with a biogenic variation in ^{13}C as plant growth preferentially favours the lighter isotope, ^{12}C .

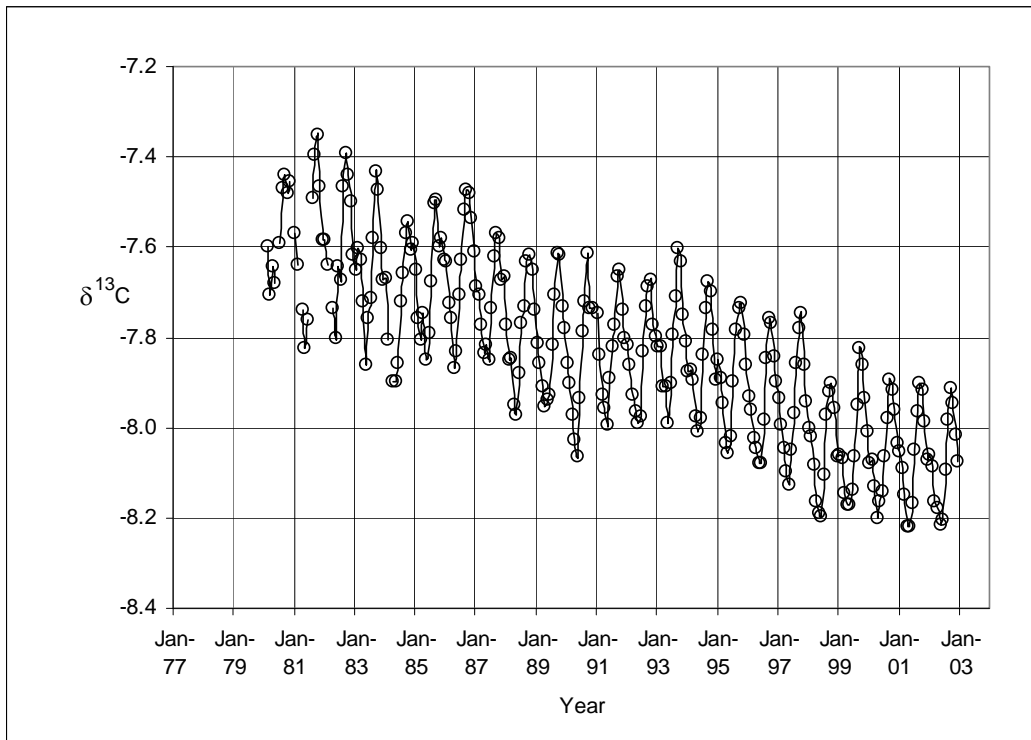


Figure 2 Monthly variations in $\delta^{13}\text{C}$ at Mauna Loa from SIO¹⁴

However at the South Pole this variation is muted and it is possible to see trends outside of the annual variations.

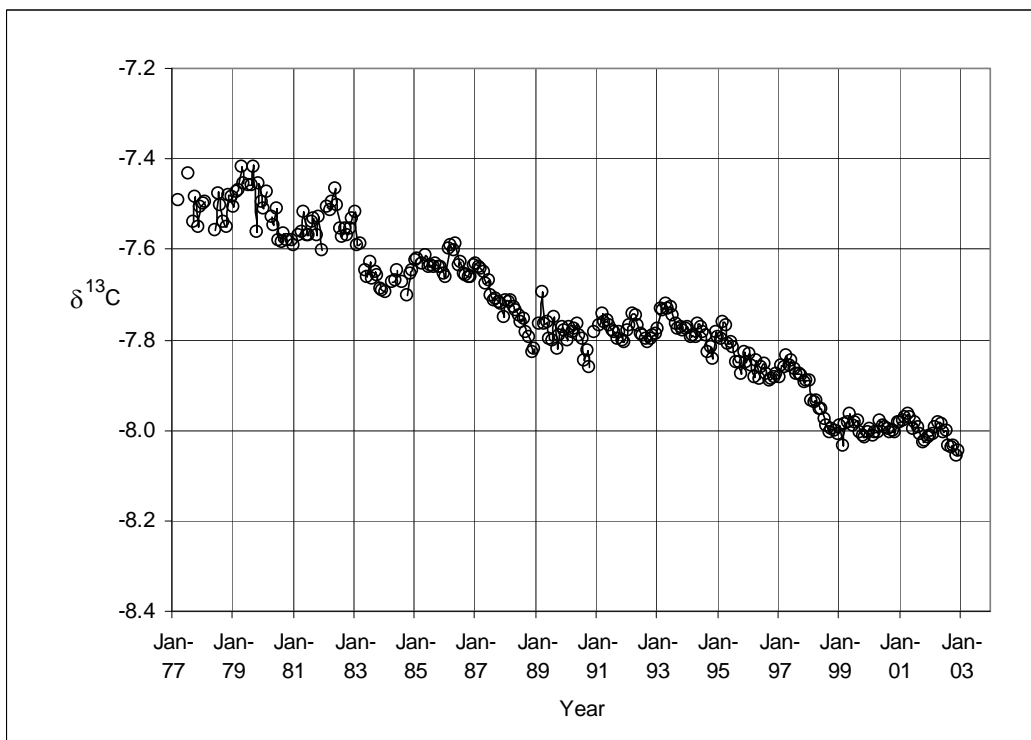


Figure 3 Monthly variations in $\delta^{13}\text{C}$ at the South Pole from SIO¹⁴

The measurements show a cascade of values with the atmosphere showing a decrease in ^{13}C . In fact the fall in ^{13}C appears as a series of steps. This behaviour is better displayed by looking at annual changes. Figure 4 shows these changes for SIO measurements from Mauna Loa and the South Pole. The annual differences in $\delta^{13}\text{C}$ are calculated from three month averages of year on year differences

of the monthly measured values in order to remove the regular seasonal cycle and to expose departures from that cycle.

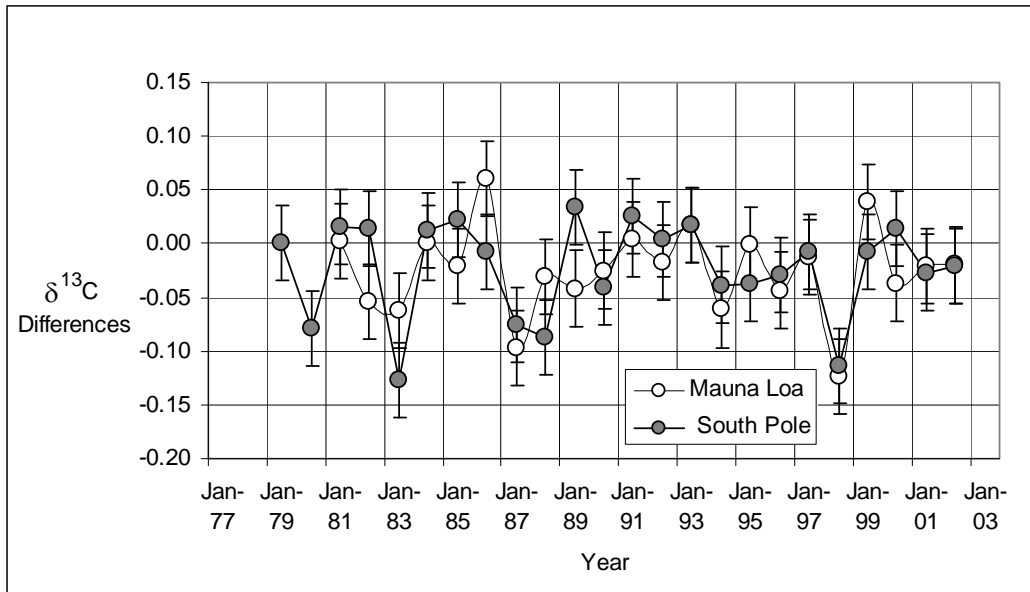


Figure 4 Three Month averages of year on year differences of the monthly measured $\delta^{13}\text{C}$ for Mauna Loa and the South Pole¹⁴.

A natural question to ask is whether these steps are associated with other variations in the atmosphere. The annual increase in CO₂ is shown in Figure 5 taking one-year differences from each location time series thus again removing a regular seasonal cycle to expose departures from that cycle.

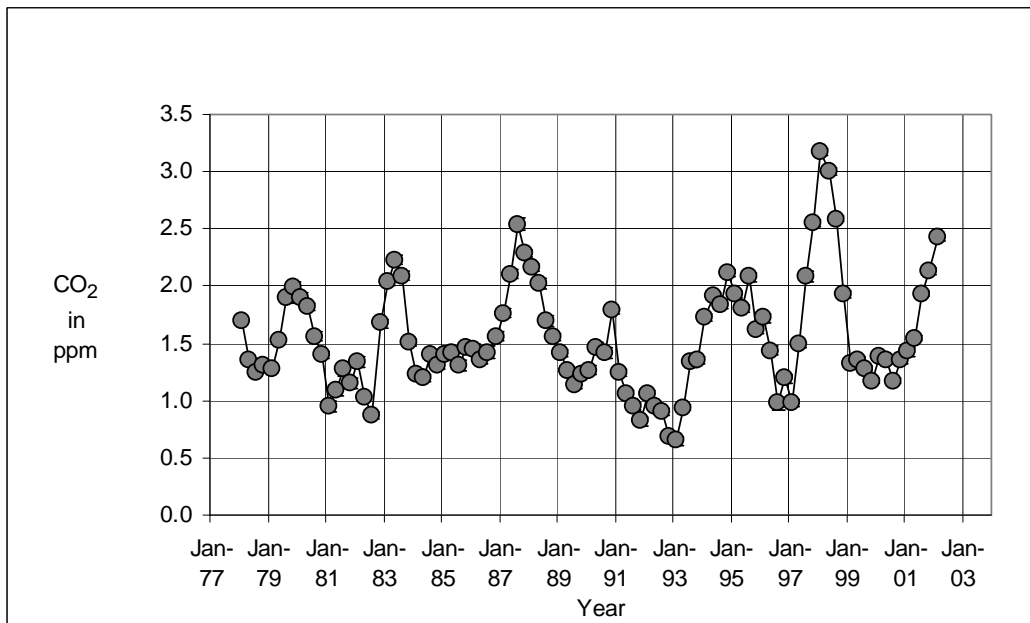


Figure 5 Three Month averages of year on year differences of the monthly measured CO₂ atmospheric concentrations for Mauna Loa and the South Pole².

Following the pattern shown in Figures 4 and 5, all 15 available data sets of ¹³C measurements have been analysed. These extend from 82°N at Alert to 90°S at the South Pole. The results are shown in Figure 6. The same differencing procedure discussed above has been applied to the changes in ¹³C

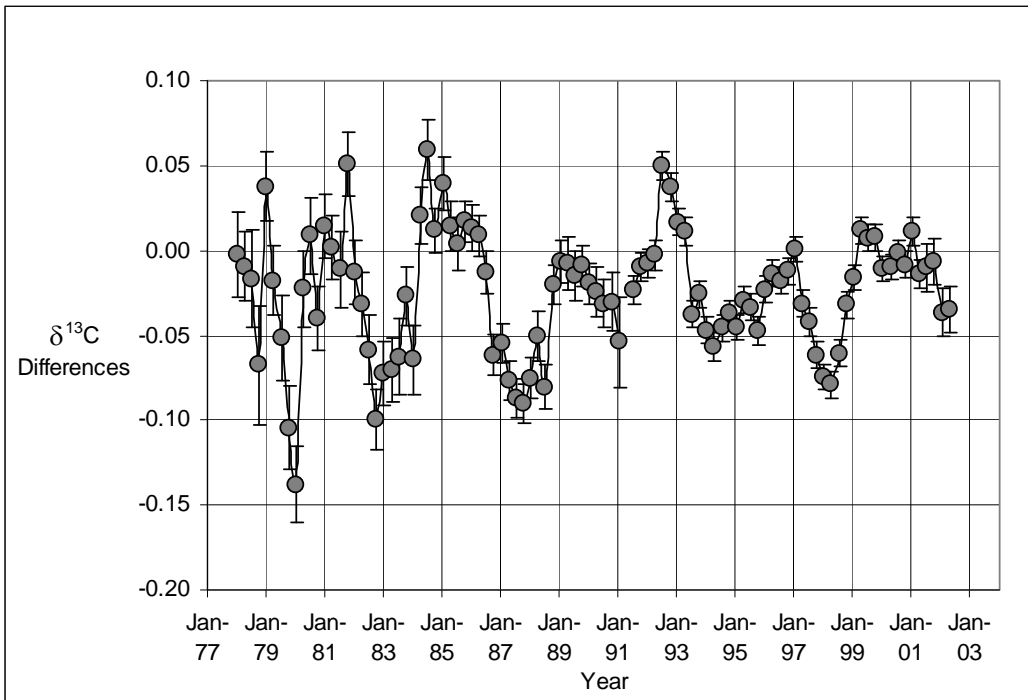


Figure 6 Three Month averages of year on year differences of the monthly measured $\delta^{13}\text{C}$ from SIO¹⁴ and CSIRO¹² data for 15 locations from Alert to the South Pole. Error bars are errors of the mean.

The location data sets have been combined on the assumption that variations in ^{13}C are simultaneous throughout the atmosphere within the accuracy of the data.

There is a discontinuity in the SIO data at the start of 1991¹⁵. The step in the SIO data and its difference to the CSIRO data is shown in Figure 7. In Figure 6 and all subsequent analysis the SIO data for 1991 has not been used.

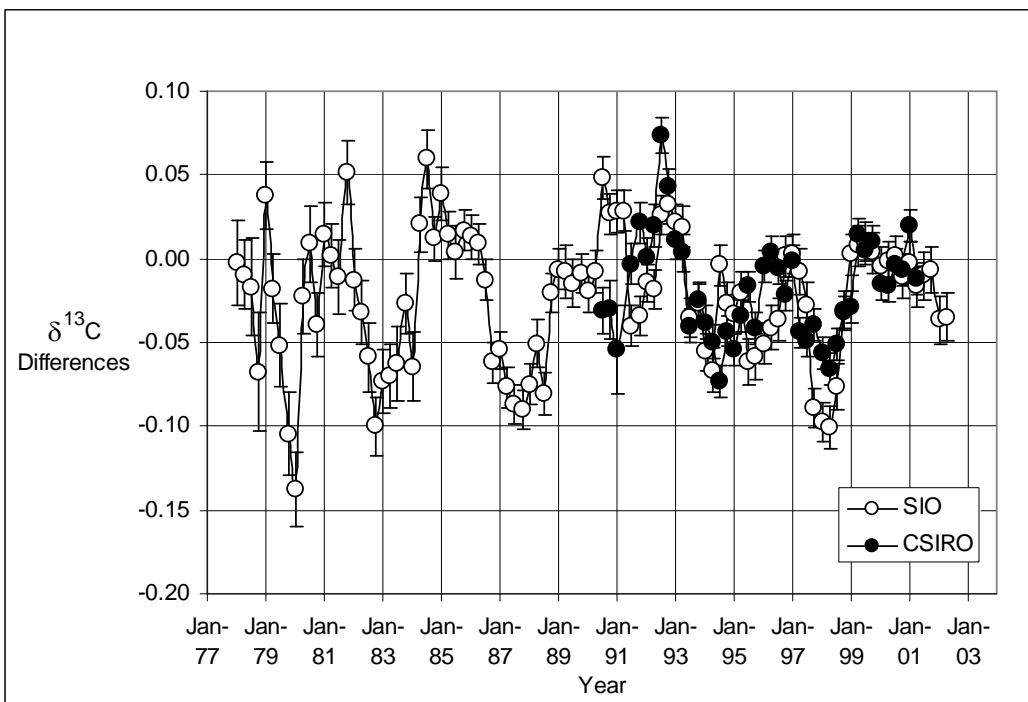


Figure 7 Separate SIO and CSIRO data series. Three Month averages of year on year differences of the monthly measured $\delta^{13}\text{C}$ from SIO¹⁴ and CSIRO¹² data for 15 locations from Alert to the South Pole. Error bars are errors of the mean.

The measuring errors combined with any time spread have been estimated by looking at the spread of measurements at each time point with from 1 to 15 locational data points. The standard deviation is found to be overall 0.050 for $\delta^{13}\text{C}$ with 0.050 for the SIO data and 0.046 for the CSIRO data. The errors plotted are the errors on the mean. Some 2,000 data points have been used in the analysis and a small number, less than 30, were rejected as being more than 4 standard deviations from the mean. This was a particular problem in the 1991-92 period where 22 SIO data points were rejected.

The correlation of CO_2 concentration with $\delta^{13}\text{C}$ gives a correlation coefficient of -0.60. Figure 8 illustrates this correlation where the $\delta^{13}\text{C}$ difference values have been plotted along with the annual changes in CO_2 concentration.

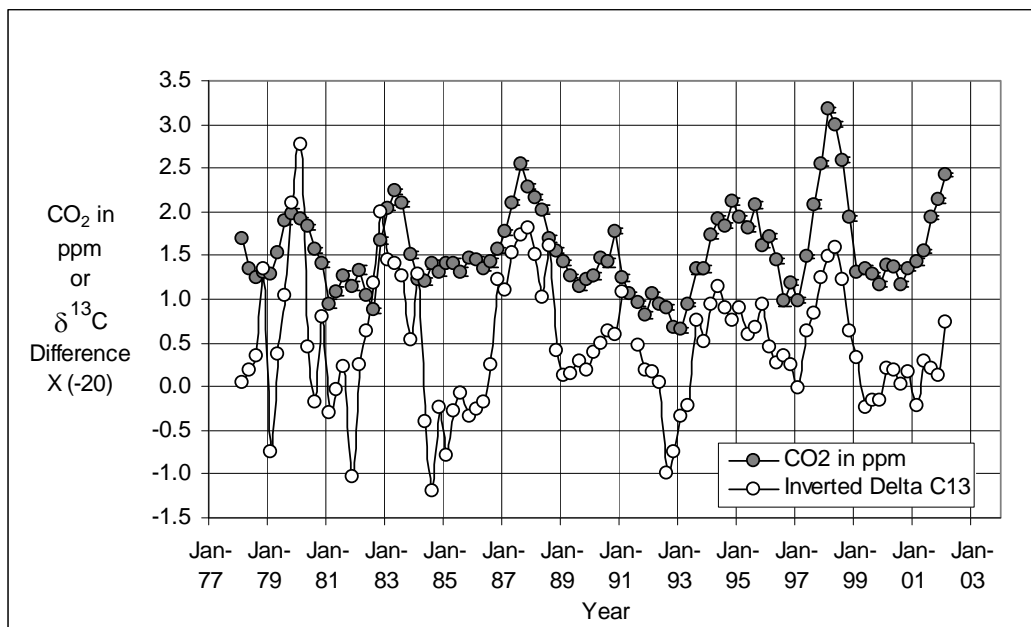


Figure 8 Correlation of CO_2 annual increases and $\delta^{13}\text{C}$ differences. The $\delta^{13}\text{C}$ differences have been inverted and scaled. Three Month averages of year on year differences of the monthly measured CO_2 concentrations for Mauna Loa and the South Pole² and $\delta^{13}\text{C}$ from SIO¹⁴ and CSIRO¹² data for 15 locations from Alert to the South Pole.

The peaks in the CO_2 concentration appear to correlate with ENSO¹⁶ events of Figure 9.

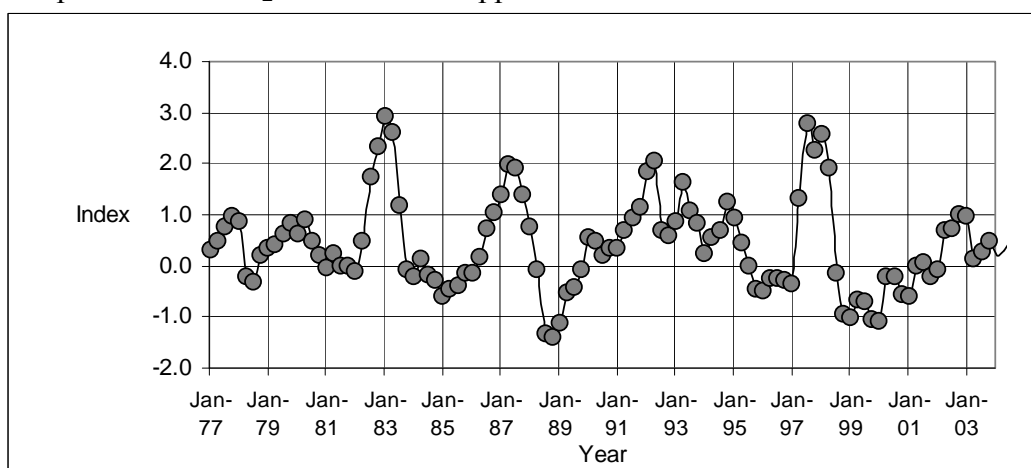


Figure 9 Multivariate ENSO Index compiled by K Walter¹⁶

So this would suggest that the change in $\delta^{13}\text{C}$ might be driven from the same source responsible for ENSO events.

3 ILLUSTRATION OF CARBON-13 CASCADE

In order to illustrate the working of the analysis, a simple set of straight lines have been matched to the South Pole data points as shown in Figure 10. The alternative of a constant addition of fossil fuel CO₂ would give a steady decrease in $\delta^{13}\text{C}$. The yearly differences (Figure 11) show the mapping of the $\delta^{13}\text{C}$ cascade.

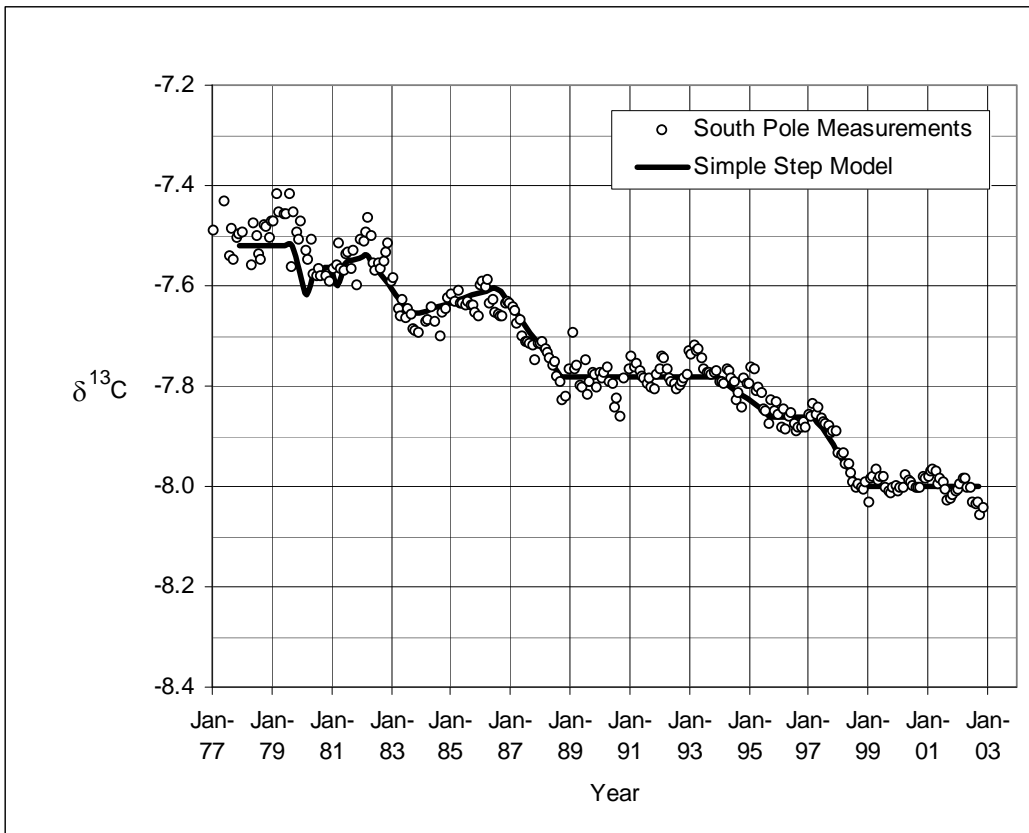


Figure 10 A simple step model of the changes in $\delta^{13}\text{C}$ at the South Pole compared to the SIO measurements.

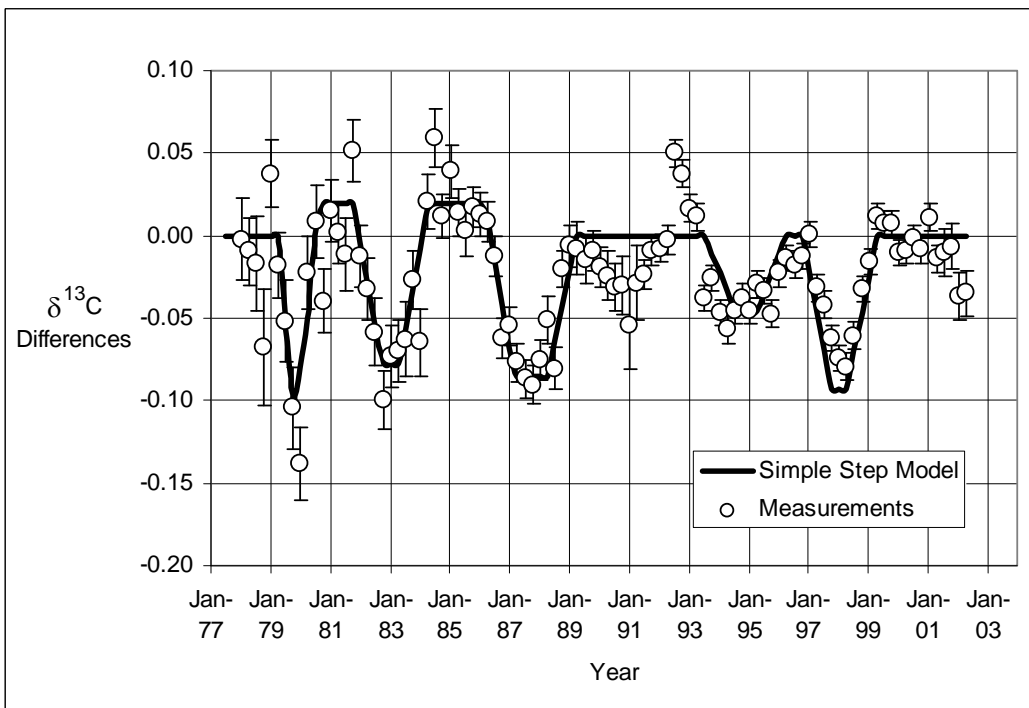


Figure 11 A simple step model of the changes in $\delta^{13}\text{C}$ at the South Pole along with the SIO measurements showing annual differences in $\delta^{13}\text{C}$.

The correlation of changes in $\delta^{13}\text{C}$ with ENSO events and the comparison with a simple model of a series of cascades suggest that the changes in $\delta^{13}\text{C}$ in the atmosphere have little to do with the input of CO_2 emissions from the continuous use of fossil fuels.

4 SEASONAL VARIATIONS OF ATMOSPHERIC CARBON DIOXIDE

The increase of CO_2 in the atmosphere is modulated by seasonal variations. These are understood to be a reflection of biogenic and chemical interactions from plants, the soil and the oceans. There is a remarkable regularity in this as can be seen in Figure 12 for the monthly measurements at Mauna Loa.

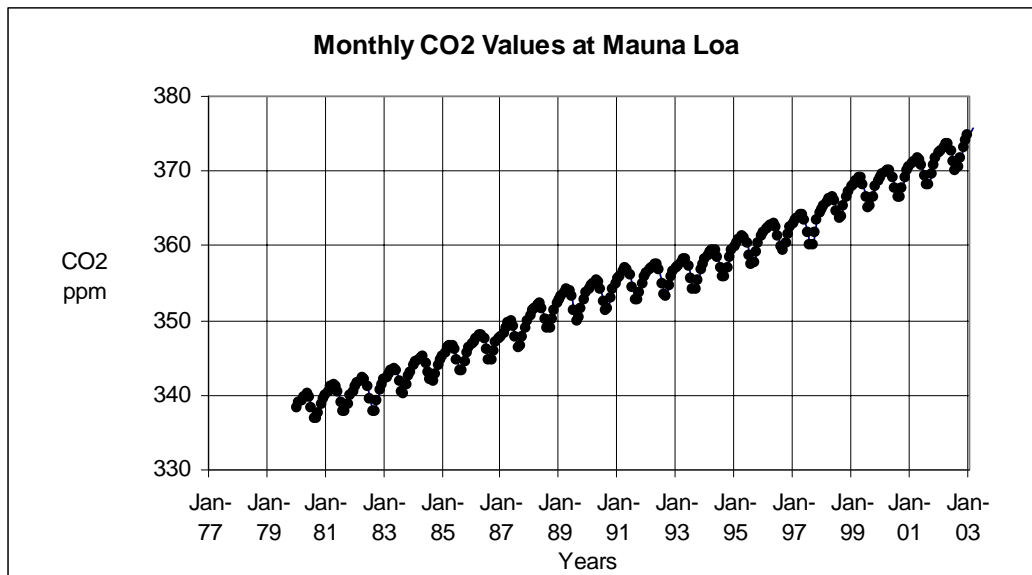


Figure 12 Monthly variations of CO_2 concentration at Mauna Loa².

The variations of CO_2 in the atmosphere over the last twenty-five years are best summarised in the following figures 13, 14 and 15 with the results listed in the Attachment. The data is from the extensive SIO and CSIRO measurements of CO_2 and from one UBA site, Schauinsland in Germany¹⁷.

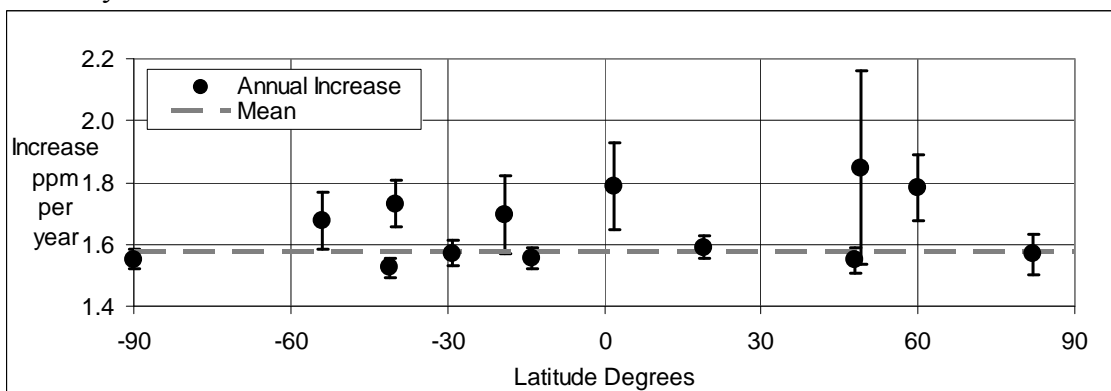


Figure 13 Annual increases in CO_2 concentration at different locations. The time span ranges from 7 to 22 years.

The annual increase of CO_2 in the atmosphere is in sharp contrast with the annual change in the seasonal variations. The mean values are:

Annual CO_2 increase	1.572 ± 0.013 ppm per year
Seasonal CO_2 increase	-0.001 ± 0.013 ppm per year

There is a difference, within errors, of a factor of 100 in annual increases.

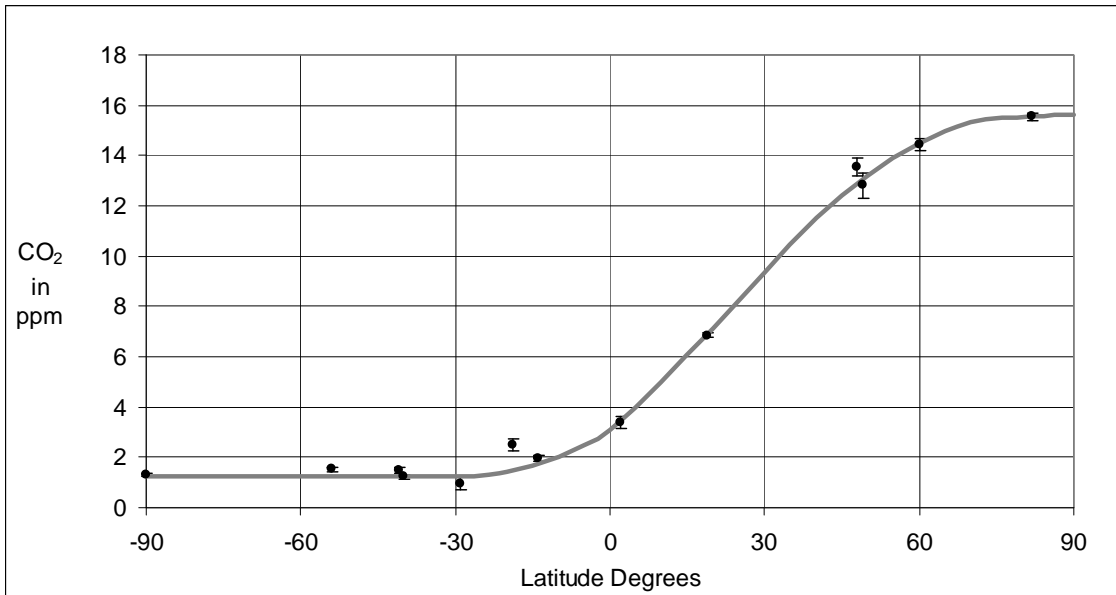


Figure 14 Seasonal variations of CO₂ concentration at different locations. The time span ranges from 7 to 22 years.

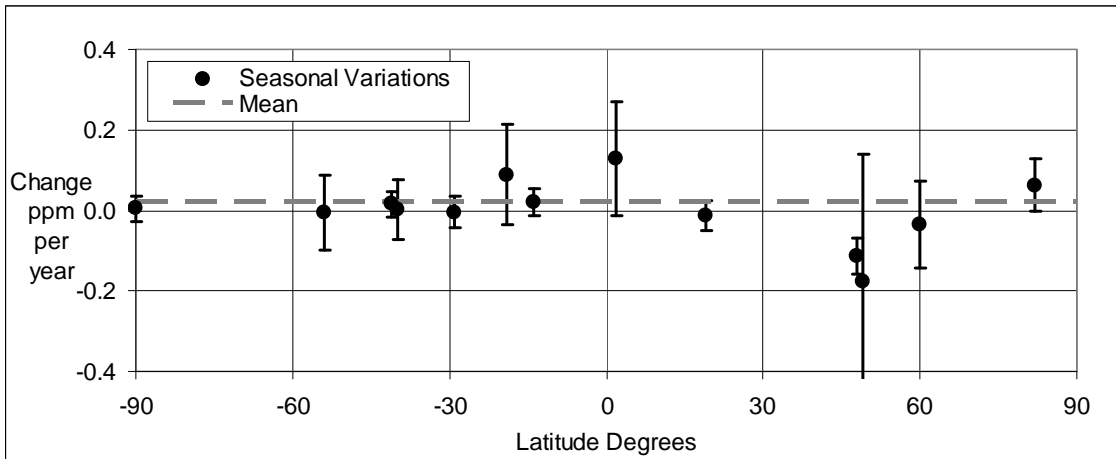


Figure 15 Annual changes in the seasonal variation of CO₂ concentration at different locations. The time span ranges from 7 to 22 years.

Over the last 20 years substantial amounts of CO₂ derived from fossil fuel have been released into the atmosphere. This has moved from 5.0 Gt of carbon in 1980 to 6.2 GtC in 1990 to 7.0 GtC in 2000. Figure 16 shows the distribution of fossil fuel CO₂ by latitude¹⁸.

Over 95% of this CO₂ has been released in the Northern Hemisphere.

To put this in context, the total carbon in CO₂ in that region of the atmosphere is some 400 GtC with seasonal net variations of 10 GtC. (This can be seen in Figure 14 where the mean seasonal net variation is 9 ppm for a total 380 ppm CO₂ concentration.)

The remarkable feature of this behaviour is that despite the accumulation in the atmosphere of fossil fuel emissions from 1980 to 2000 at the rate 3 GtC per year, there has been virtually no change in the seasonal variations.

If the rise in CO₂ concentration is understood to be from fossil fuel CO₂ but the balance of emissions is taken up by sinks of CO₂ then it follows that there is an increase in the capacity of the

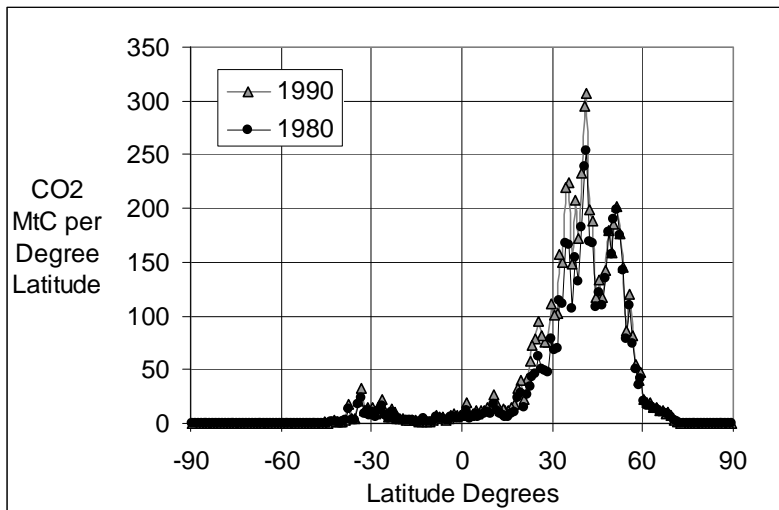


Figure 16 Estimated Atmospheric CO₂ from fossil fuels. Source CDIAC¹⁸.

sinks to absorb CO₂. If this is the case then one might expect changes in the seasonal variations in proportion to the expansion of the land-based or biogenic sinks of CO₂.

The estimate of the effects of added CO₂ is shown for the Northern Hemisphere in Table 1. The carbon cycle data is taken from the 3rd IPCC Report¹⁹.

Carbon Cycle			Atmosphere		CO ₂ from Fossil Fuel	
			1980	2000	1980	2000
Global						
Atmospheric Concentration of CO ₂	ppm		350	381		
Increase per year	ppm	1.572				
Amount of CO ₂ in Atmosphere Tonnes	GtC		750.0	816.4		
Total Atmosphere CO ₂ Exchanges	GtC	28%	210.0	228.6		
Atmosphere-Land CO ₂ Exchanges	GtC	16%	120.0	130.6		
Increase per year	GtC/year		3.4	3.4	5.30	7.00
Northern Hemisphere						
Atmosphere Fraction		50%				
Amount of CO ₂ Tonnes	GtC		375.0	408.2		
Atmosphere-Land CO ₂ Exchanges	GtC	22%	80.6	87.8		
Net Seasonal Variations	ppm		9.4	9.4		
~12% of land exchanges	GtC		10.1	10.1		
Fossil Fuel CO₂ Emitted						
Land source/sink	GtC	100%			5.30	7.00
Emitted CO ₂ absorbed by land sink	GtC	30%			1.59	2.10
Increase to Seasonal Variations	GtC				0.20	0.24
~12% of additional absorbed CO ₂						
Estimated Increase per year	ppm				0.185	0.225
Seasonal Variations						
Measured Increase per year	ppm				0.013	0.013
Seasonal Variations						
(-0.001+/- 0.013)						

Table 1 Estimate of expected increase in seasonal variations for CO₂ in the Northern Hemisphere. The third column shows the seasonal variations for 1980 and 2000. The fourth column shows the estimated contribution from the continuous production of fossil fuel CO₂ for 1980 and 2000. The CO₂ from fossil fuel is assumed to be distributed 50% to the atmosphere, 20% to the ocean and 30% to biogenic sinks. In the Northern Hemisphere the land exchange has been increased as there is 40% land cover compared to 20% for the Southern Hemisphere.

The calculations in the tables are order of magnitude estimates of the variations that might be expected if 30% of the fossil fuel sourced CO₂ was taken up in biogenic sinks.

In addition there is a 10% seasonal variation in CO₂ from fossil fuel use in North America and a similar behaviour might be expected in Europe. These variations have not been considered.

The estimates indicate limits to the variations that are substantially larger than the measured values. This assumes that the measured value is independent of latitude.

It is remarkable that emitted CO₂ is thought to be partially absorbed by CO₂ sinks but yet leaves no hint of expected biogenic variation. Increasing CO₂ might be expected to encourage plant growth. A simple conclusion that might be drawn from this is that the fossil fuel derived CO₂ is almost totally absorbed locally in the year that it is emitted. An alternative explanation would require the ocean to be the sink for CO₂. The absence of change remains a puzzle.

5 NORTH-SOUTH HEMISPHERE TIMING DIFFERENCES

Another interesting approach is to see if there are timing differences between the Northern and Southern Hemispheres (NH and SH) with the emission of CO₂.

A tracer for CO₂ transport from the NH to the SH was provided by ¹⁴C created by nuclear weapons testing in the 1950's and 1960's. The analysis of ¹⁴C in atmospheric CO₂^{20, 21} shows in Figure 17 that it took some years for exchanges of CO₂ between the hemispheres before the ¹⁴C was uniformly distributed.

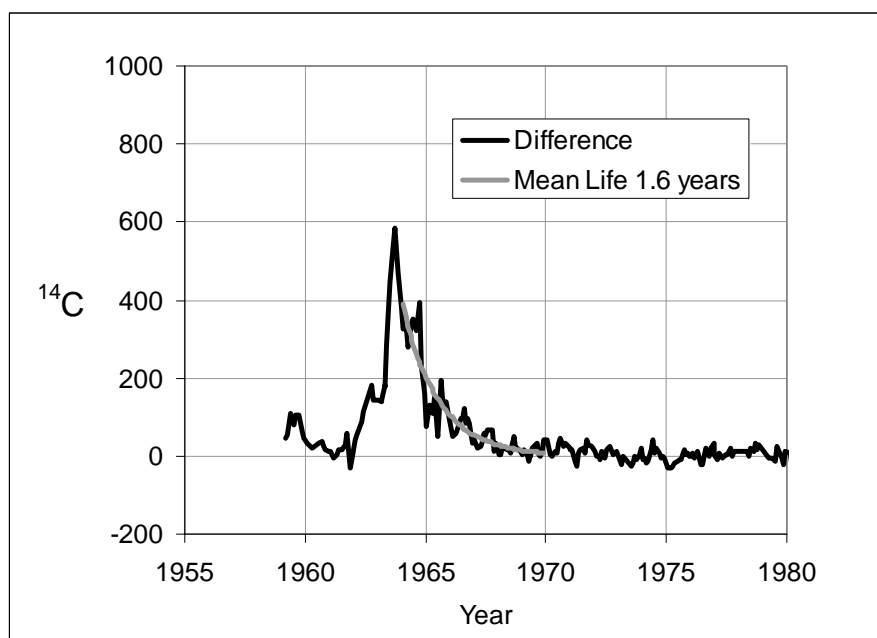


Figure 17 Timing differences of Northern and Southern Hemisphere carbon pulse. A number of massive weapons were detonated in the Arctic circle just before the Partial Test Ban Treaty came into force in October 1963. The measurements were made in New Zealand²⁰ and Germany²¹.

If 75% of CO₂ from fossil fuel is emitted north of latitude 30 then some time lag might be expected due to the sharp year-to-year variations in the estimated amounts left in the atmosphere. A simple model, following the example of the ¹⁴C data with a one year mixing time, would suggest a delay of

6 months for CO₂ changes in concentration in the Northern Hemisphere to appear in the Southern Hemisphere.

A correlation plot of SIO year on year differences of monthly measurements at Mauna Loa against those at the South Pole is shown in Figure 18. The time difference is positive when the South Pole data leads the Mauna Loa data. Any negative bias (asymmetry in the plot) would indicate a delayed arrival of CO₂ in the Southern Hemisphere.

There does not appear to be any time difference between the hemispheres. This suggests that the annual increases may be coming from a global or equatorial source.

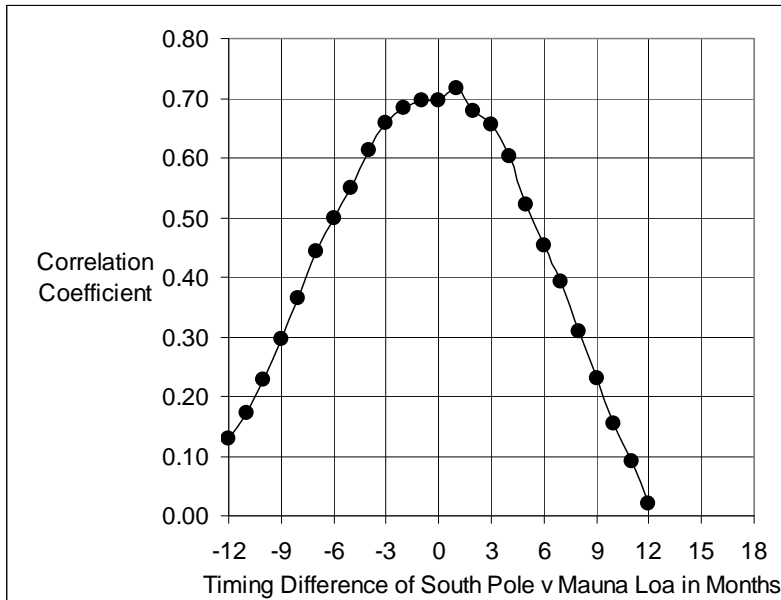


Figure 18 Variation in the correlation coefficient between year on year differences in measured monthly CO₂ concentrations at Mauna Loa and the South Pole as the times series are separated. A negative timing bias would indicate the delayed arrival of CO₂ in the Southern Hemisphere.

A similar comparison has been made for the variations of $\delta^{13}\text{C}$ shown in Figure 19. The data has been analysed as 3-month averages. The result is not straightforward and suggests possible variations in the two hemispheres with time delays in both directions.

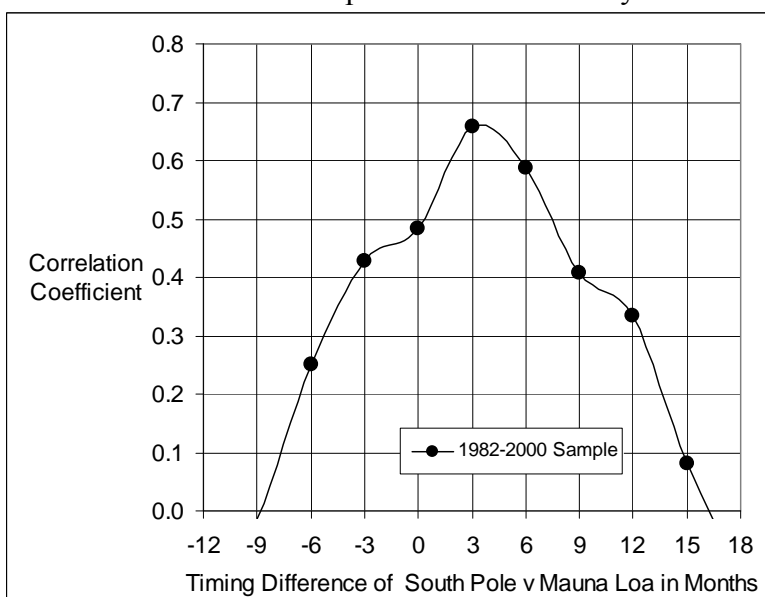


Figure 19 Variation in the correlation coefficient between year on year differences of three monthly average $\delta^{13}\text{C}$ measurements at Mauna Loa and the South Pole as the times series are separated. A positive timing bias indicates the delayed arrival of CO₂ in the Northern Hemisphere.

If the increases in atmospheric CO₂ are being driven by fossil fuel emissions then changes in the isotope ratio would be expected to occur first in the Northern Hemisphere and then move to the Southern Hemisphere. The result suggests that the changes in the isotope ratio start in the Southern Hemisphere. This suggests an equatorial or Southern source of CO₂ emission.

6 CONCLUSION

During the 1977 to 2001 time period analysed:

Changes in the isotope ratio are discontinuous. The temporal peaks in ¹³C appear to correlate with the CO₂ concentration changes. Further the temporal peaks in ¹³C and the CO₂ peaks correlate with ENSO events.

The yearly increases of atmospheric CO₂ concentrations have been nearly two orders of magnitude greater than the change to seasonal variation which implies that the fossil fuel derived CO₂ is almost totally absorbed locally in the year that it is emitted.

A time comparison of the SIO measurements of CO₂ at Mauna Loa with the South Pole shows a lack of time delay for CO₂ variations between the hemispheres that suggests a global or equatorial source of increasing CO₂. The time comparison of ¹³C measurements suggest the Southern Hemisphere is the source. This does not favour the fossil fuel emissions of the Northern Hemisphere being responsible for their observed increases.

All three approaches suggest that the increase of CO₂ in the atmosphere may not be from the CO₂ derived from fossil fuels. The ¹³C data is the most striking result and the other two approaches simply support the conclusion of the first approach.

It is not obvious that the pulsed contributions of CO₂ are related to continuous fossil fuel emissions given the seasonal data and the lack of a time delay in the Southern Hemisphere. Could it be a biogenic signal? After all the Northern Hemisphere has seasonal fluctuations of up to 15 ppm so could a 2 ppm pulse be related to changing conditions in tropical lands as a result of El Nino's?

The conclusion is that CO₂ emissions from fossil fuels do not make it to the "well mixed" atmosphere but are probably fixed locally. The increase in CO₂ is driven by other processes related to the natural variability of the climate. Some of this CO₂ variability is correlated with and may be related to ENSO events.

Acknowledgements

The author wishes to thank W. Kininmonth, R. Carter and G. Paltridge for many extensive tutorials. In addition T. Segalstad and J. McLean have supplied valuable references and help on particular sections of the paper.

Attachment: Table of Data used in Figures 13 to 15

Location and Latitude	No. of Years	Annual Increase of CO ₂ ppm per year		Seasonal Variations of CO ₂ ppm		Annual Change of Seasonal Variations ppm per year		
		Value	Error	Value	Error	Value	Error	
South Pole	-90	22	1.551	0.031	1.32	0.05	0.005	0.031
Macquarie Island	-54	9	1.676	0.093	1.52	0.09	-0.006	0.093
Baring Head	-41	27	1.524	0.031	1.46	0.12	0.015	0.031
Cape Grim	-40	9	1.731	0.075	1.23	0.08	0.003	0.075
Kermadec	-29	22	1.571	0.040	0.97	0.26	-0.005	0.040
Cape Ferguson	-19	10	1.696	0.124	2.49	0.22	0.089	0.124
American Samoa	-14	22	1.557	0.034	1.96	0.10	0.020	0.034
Christmas Island	2	9	1.788	0.140	3.38	0.23	0.128	0.140
Mauna Loa	19	22	1.591	0.037	6.85	0.07	-0.013	0.037
Schauinsland	48	20	1.548	0.042	13.57	0.36	-0.113	0.046
Point Estevan	49	7	1.848	0.314	12.81	0.50	-0.175	0.314
Shetland Islands	60	9	1.783	0.106	14.45	0.23	-0.035	0.106
Alert	82	19	1.569	0.065	15.54	0.15	0.062	0.065
Mean Values			1.572	0.013			-0.001	0.013

Table Results of straight line fits to quantities derived from monthly measurements of CO₂ concentrations from SIO² and CSIRO⁸ data sources.

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- ¹ See for instance IPCC Climate Change 2007, Synthesis Report, Summary for Policymakers
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