AN EMPIRICAL STUDY OF FOSSIL FUEL EMISSIONS AND OCEAN ACIDIFICATION

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ABSTRACT: Detrended correlation analysis of annual fossil fuel emissions and mean annual changes in ocean CO2 concentration in the sample period 1958-2014 shows no evidence that the two series are causally related. The finding is inconsistent with the claim that fossil fuel emissions have a measurable impact on the CO2 concentration of the oceans at a lag and time scale of one year. The results are presented with the disclaimer that the CO2 data time series is discontinuous.[1]

1. INTRODUCTION

Environmentalists and climate scientists have for long struggled with the issue of fossil fuel emissions because this source of carbon appears to be external and unnatural and foreign to the surface biota, the natural carbon cycle, and to a fragile climate and environmental equilibrium that nurtures life on earth. It is thought that the "extra" CO2 collects in the planet's CO2 inventories either in the atmosphere where its accumulation can cause dangerous climate change by way of the greenhouse effect and global warming (Callendar, 1938) (Hansen, 1984) (IPCC, 2014), or in the oceans where its unnatural excess can have harmful effects on marine life by way of ocean acidification (Caldeira K. , 2003) (Doney, 2004) (McNeil, 2006). In both cases the time scale of these effects, that is the time from emission to a measurable change in accumulation, is thought to be annual (NOAA-1, 2015) (Scripps, 2013) (IPCC, 2007) (IPCC, 2014). In previous works we looked at the carbon flow accounting methodology that leads to the accumulation hypothesis (Munshi, Uncertain flow accounting, 2015) and presented a critical evaluation of the link between fossil fuel emissions and changes in atmospheric composition (Munshi, Responsiveness of Atmospheric CH4, 2015) (Munshi, Responsiveness of atmospheric CO2, 2015). In this short note we look at empirical evidence that could support a causal link between fossil fuel emissions and ocean acidification.

The oceans form the largest known reservoir of inorganic CO2 on the surface of the planet. As such it plays an important role in the natural carbon cycle and in the 'carbon budget' of the surface-atmosphere climate system (IPCC, 2014). According to the IPCC, the inorganic dissolved CO2 content of the oceans, not including the ocean floor, was 38,000 GTC (gigatons of carbon equivalent) in 2009 (IPCC, 2014). Using a figure of 1.35×10^{21} liters as the volume of the oceans, the equivalent CO2 concentration of the oceans is 2.345 mmol/L (millimoles per liter) in close agreement with more than 124,000 measurements made by oceanic research vessels since 1958 that show an average of 2.244 mmol/L. This work adds to the rich and growing body of investigation into the impact of fossil fuel emissions on ocean acidification by presenting this data in relation to fossil fuel emissions as an empirical study to complement prior works that relied mostly on climate models and laboratory experiments (Cornwall, 2015) (Caldeira K. , 2003) (McNeil, 2006) (Flynn, 2015) (Munday, 2011).

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[1] Date: October 2015, updated 10/8/2015 to include minor changes requested by Bruce Keele

Key words and phrases: ocean acidification, global warming, climate change, Monte Carlo simulation, anthropogenic emissions, carbon dioxide, detrended analysis, human emissions, fossil fuels, bootstrap, numerical methods, natural variability

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2. DATA AND METHODS

The National Oceanic and Atmospheric Administration of the United States maintains a large repository of data on the world's oceans called the World Ocean Database (NOAA-2, 2015). This database contains a detailed data record of more than one hundred oceanic research cruises that set a total of 7,762 measurement anchors from 1958 to 2014. Of these anchor positions, 3,985 (51.4%) were in the Northern Hemisphere, 3,765 were in the Southern Hemisphere, and the remaining 12 measurement anchors were exactly on the equator. The longitudinal distribution of the measurement anchors show 5,555 (71.6%) to be west of the Prime Meridian and 2,205 (28.4%) to be east of the Prime Meridian. The remaining 2 measurement anchor locations were exactly 180 degrees from the Prime Meridian.

At each anchor position, temperature and composition measurements were taken at depths of 5 meters down to depths of more than a kilometer in steps of 20 meters or more. At each depth the temperature is recorded in degrees Centigrade and the CO2 concentration in mmol/L. On average, measurements are made at 15 to 25 depths per location. A sum total of 128,109 values of CO2 concentration, along with the corresponding temperature, are reported. Data records containing values of CO2 concentrations less than 1 mmol/L or greater than 4 mmol/L were identified as anomalous. A total of 3,296 data records were removed from the dataset on this basis. The remaining 124,813 observations form the dataset for this empirical investigation of ocean acidification. A screenshot of a portion of the dataset is shown in Figure 1. The sequence has been scrambled so that a variety of data may be made visible. The mean, median, standard deviation, and range of each variable are displayed. The DNO is an ordinal depth designation. Depth is reported in meters and CO2 in mmol/L.

Figure 1: Marine CO2 concentration dataset

	A	B	C	D	E
1	MIN	1	0	-2.31383	1.015
2	MAX	60	6453.5	31.3417	2.8765
3	MEDIAN	13	765.56	4.156	2.24372
4	MEAN	14.714	1313.105	7.691	2.244
5	STDEV	9.595	1406.428	7.776	0.110
6	YEAR	DNO	DEPTH	DEGC	CO2
7	1986	13	690	8.45	2.343
8	1996	12	4671	1.16	2.328
9	2002	6	100	1.05	2.195
10	1997	9	550	8.53	2.193
11	2013	24	3625	2.39	2.251
12	2009	3	52	20.79	2.090
13	1997	3	44	25.61	2.007
14	2010	35	3070	1.78	2.351
15	2011	33	3631	0.33	2.319
16	2009	11	248	14.31	2.227
17	2003	17	1385	3.00	2.300

A peculiarity of this dataset is the non-homogeneity of the data across the sample period of 1958-2014 (Table 1). In the 57-year period we find 23 years with no data available. The remaining 34 years are unequally represented in terms of the number of measurements available.

Year	Mean	Stdev	Count
1958	2.084	0.104	301
1959	2.130	0.066	154
1960	2.111	0.112	181
1961			0
1962			0
1963			0
1964	2.173	0.109	319
1965	2.119	0.111	931
1966	2.162	0.120	1353
1967			0
1968			0
1969			0
1970			0
1971			0
1972			0
1973			0
1974			0
1975			0
1976			0
1977			0
1978			0
1979			0
1980			0
1981			0
1982			0
1983			0
1984			0
1985	2.240	0.092	625
1986	2.271	0.122	776
1987	2.269	0.070	720
1988			0
1989	2.230	0.069	352
1990	1.929	0.139	117
1991	2.247	0.139	1866
1992	2.188	0.126	2557
1993	2.242	0.101	6480
1994	2.258	0.115	10816
1995	2.214	0.104	3732
1996	2.234	0.105	4819
1997	2.201	0.063	6171
1998	2.205	0.051	3273
1999	2.232	0.097	3998
2000			0
2001	2.270	0.122	2217
2002	2.195	0.031	1433
2003	2.228	0.085	9554
2004	2.259	0.120	1199
2005	2.265	0.113	6602
2006	2.283	0.142	2852
2007	2.312	0.126	9030
2008	2.235	0.119	7146
2009	2.238	0.104	10069
2010	2.238	0.099	2823
2011	2.275	0.075	6316
2012	2.245	0.113	4803
2013	2.259	0.116	6591
2014	2.254	0.077	4631

Table 1: The time heterogeneity of the CO2 data

The important response variable in this empirical study is the annual change in the CO2 concentration. The gaps in the time series greatly reduce the number of annual differences that can be studied. In a 57-year study period, only 27 year to year differences in CO2 can be computed and because of large

differences in sample size, the number of differences that may be computed is limited to the smaller of the two sample sizes.

The uncertainty in the annual means of the CO2 data are computed as the standard deviation of all measurements reported for that year. Some of the differences within a given year of course arise from differences in temperature The analysis is therefore carried out twice, once with the raw values of CO2 as measured regardless of temperature, and again with all CO2 values set to an equivalent value at the a standard temperature of 25°C or 298.15^{o}K. The conversion of CO2 from temperature T to the equivalent concentration at standard temperature S=25°C is carried out as CO2(S) = CO2(T)*298.15/(T+273.15).

The data for carbon emissions from fossil fuels and cement manufacture (Boden, 2013) are provided by the Carbon Dioxide Information Analysis Center of the Oak Ridge National Laboratories (CDIAC, 2015) on an annual basis from 1750 to 2010. The data are provided as millions of metric tons of carbon equivalent and they are converted to GTC (gigatons of carbon equivalent). The value for 2011 was taken from the IPCC AR5 Chapter 6 as 9.5 GTC (IPCC, 2014). Emission values for 2012, 2013, and 2014 were derived from estimates provided by the Global Carbon Project (GCP, 2015). The emissions data used in this study is thus a continuous annual time series from 1958 to 2014. The CDIAC does not provide uncertainty information for the data but both the IPCC (IPCC, 2014) and the Global Carbon Project (GCP, 2015) indicate that the uncertainty in fossil fuel and cement emission values (emissions[2]) should be set to a standard deviation equal to five percent of the mean.

Emissions from land use changes are not used in this study. They are irrelevant to the specific point raised by all theorists from Callendar to the IPCC that the theory of climate system perturbation has to do with the introduction of carbon from sources that were previously insulated from the ocean-atmosphere system. From an empirical standpoint, fossil fuel emissions provide better statistical power since the land use change emissions are rather constant over the years and with a much larger degree of uncertainty.

The analysis is carried out with annual mean CO2 values because that is the only way that year to year differences can be computed. All graphic displays are based on this relatively small number of annual means. A weakness of this method is that the statistical power of pooling the variances of the annual means is foregone in the analysis of annual means. A Monte Carlo bootstrap procedure is used to address these concerns. The procedure creates a synthetic large sample of annual differences that are true to the sample size and uncertainty of the original data. The synthetic large samples are then used to check the results of the analysis of annual means. There are three parameters in the analysis methodology - annual means versus large sample, whether CO2 measurements are corrected for temperature, and whether uncertainty is taken into account.

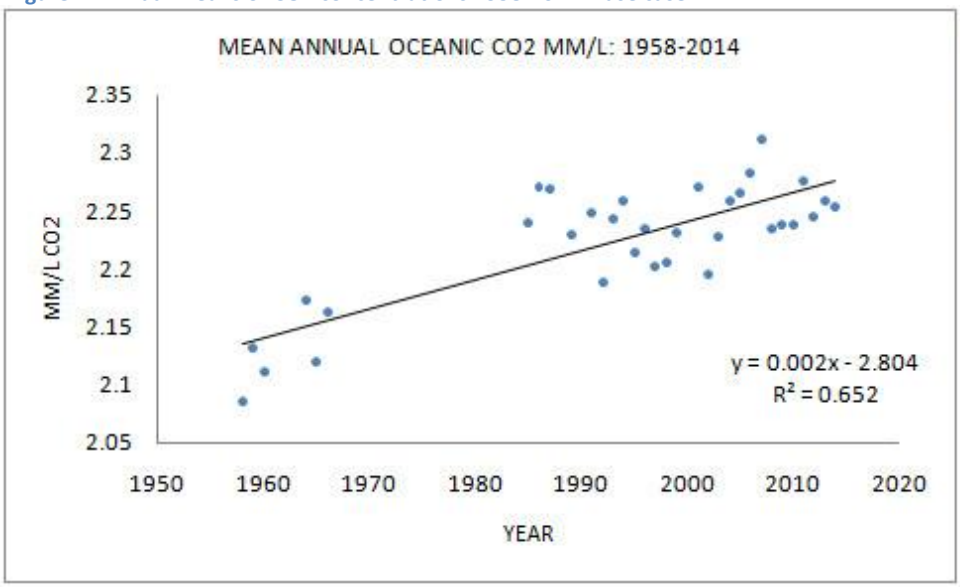
All data and computational details used in this study are available in an online data archive (Munshi, Acidification Paper Archive, 2015).

[2] Henceforth the word emissions will be used to mean emissions from fossil fuel combustion and cement manufacture.

3. DATA ANALYSIS

3.1 Base case: Annual means, no temperature correction, equally weighted, no uncertainty

Figure 2: Annual means of CO2 concentrations 1958-2014: Base case



REGRESSION RESULTS

0.00252	-2.80481
0.00033	0.65966
0.65208	0.03183
t-stat=	7.62
p-val=	0.00000

Figure 2 displays the annual mean CO2 data graphically. Note the discontinuities. The graph indicates a rising trend in oceanic CO2 concentration as measured and without consideration for temperature[3]. Using the regression statistics shown we can carry out a hypothesis test for

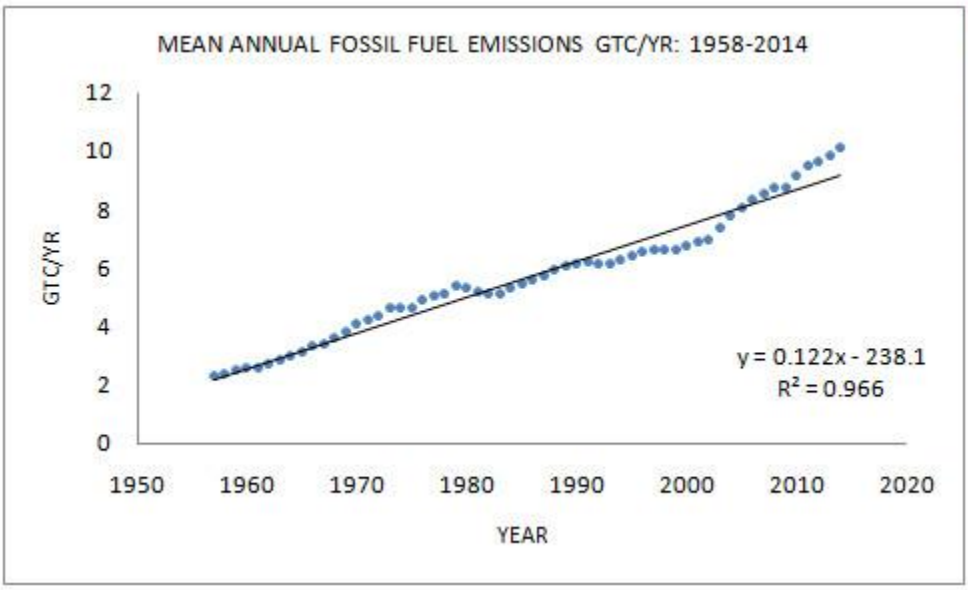
H_0: $\beta=0$ against H_A: $\beta\neq0$ with $\alpha=0.001$

Since the p-value < α, we reject H_0 and conclude that oceanic CO2 concentrations have been rising in this period. The low value of the maximum false positive error rate of $\alpha=0.001$ is consistent with "Revised standards for statistical evidence" published by the National Academy of Sciences to address an unacceptably high rate of irreproducible results in published research (Johnson, 2013). When multiple comparisons are made the value of α per comparison will be set to a lower value in accordance with the Bonferroni adjustment procedure (Holm, 1979).

The question we wish to address is whether the observed rise in oceanic CO2 can be ascribed to fossil fuel emissions. The response time for the proposed causal relationship between emissions and oceanic CO2 is widely held to be one year (NOAA-1, 2015) (Scripps, 2013). For the empirical test, we take that to mean for example that emissions in the calendar year 1958 can be related to the difference in mean annual oceanic CO2 between 1959 and 1958. The data for annual fossil fuel emissions and corresponding annual changes in oceanic CO2 are shown graphically in Figure 3 and Figure 4..

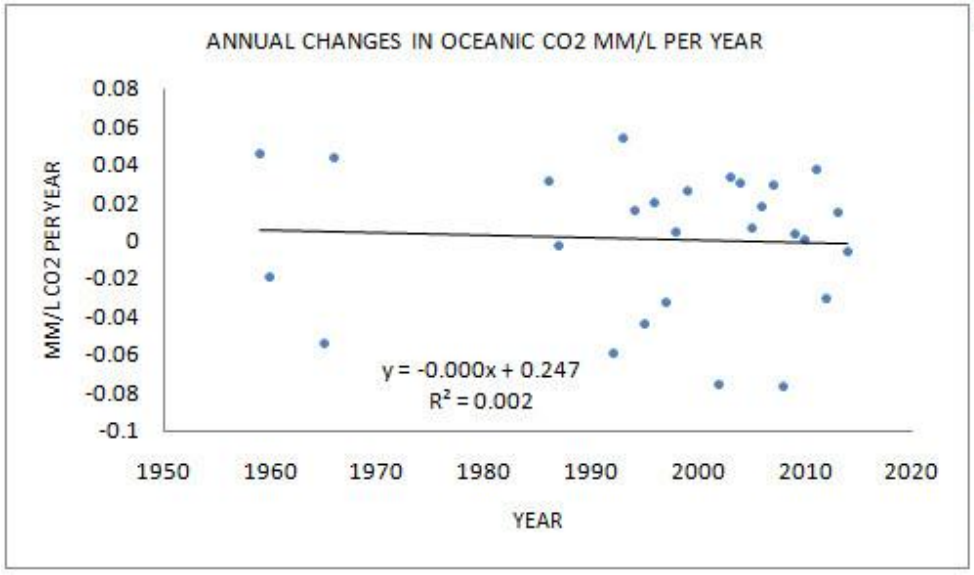
[3] CO2 concentrations are lower at higher temperatures ceteris paribus and this relationship is addressed in the next section.

Figure 3: Annual emissions in GTC per year 1958-2014



REGRESSION RESULTS	
0.12283	-238.18
0.00303	6.02558
0.96695	0.38690
t-stat=	40.47
p-val=	0.00000

Figure 4: Annual changes in oceanic CO2 1959-2014



REGRESSION RESULTS	
-0.00012	0.247718
0.000463	0.924735
0.002853	0.038218
t-stat=	0.27
p-val=	0.79086

We can see in these figures that the strong rising trend in emissions is not matched by changes in CO2. A rigorous test for this intuition may be carried out with a hypothesis test for the correlation between the two time series after they have been detrended. Detrending ensures that the effect being measured is an annual frequency response and not an artifact of a common long term drift in time (Box, 1994) (Podobnik, 2008) (Watkins, 2011) (Shumway, 2011) (Santer, 2011). The two detrended series are shown graphically in Figure 5 and the correlation between them is assessed with a hypothesis test in Figure 6. The results do not indicate that there is a correlation between the annual rate of fossil fuel emissions and annual changes in oceanic CO2 concentration. The evidence does not support the hypothesis that human emissions cause ocean acidification on an annual time scale. All data and computational details are available in the online data archive for this paper (Munshi, Acidification Paper Archive, 2015).

Figure 5: The detrended series

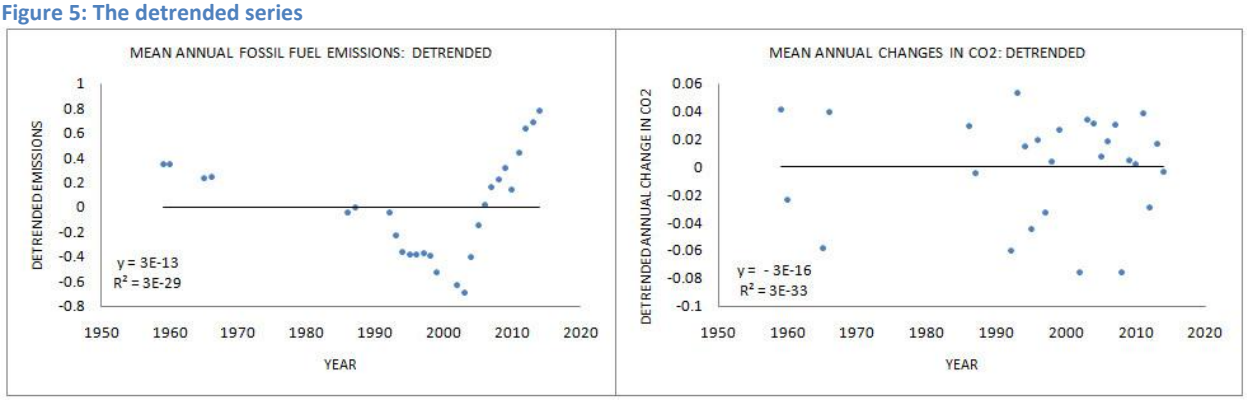
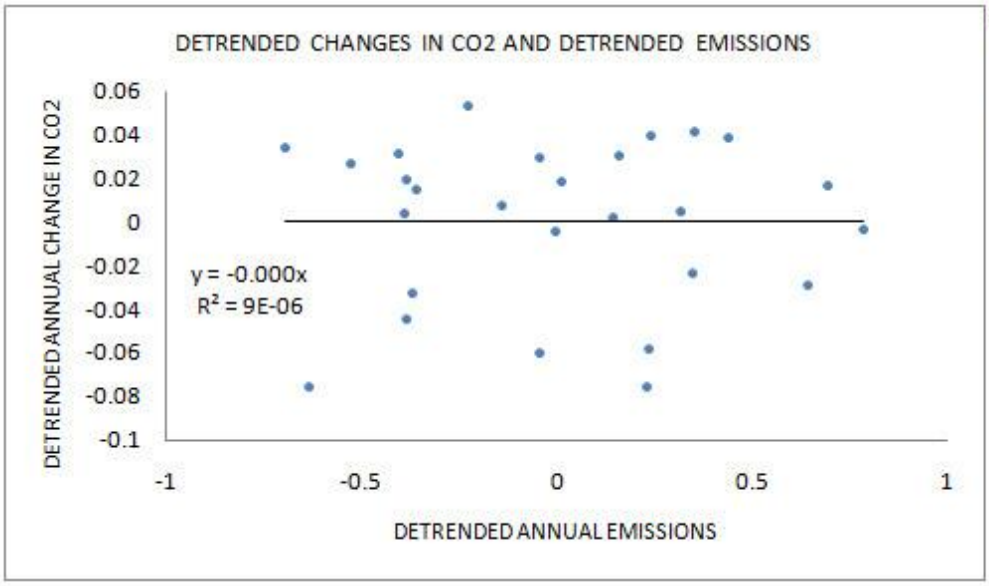


Figure 6: The correlation between the detrended series



REGRESSION RESULTS

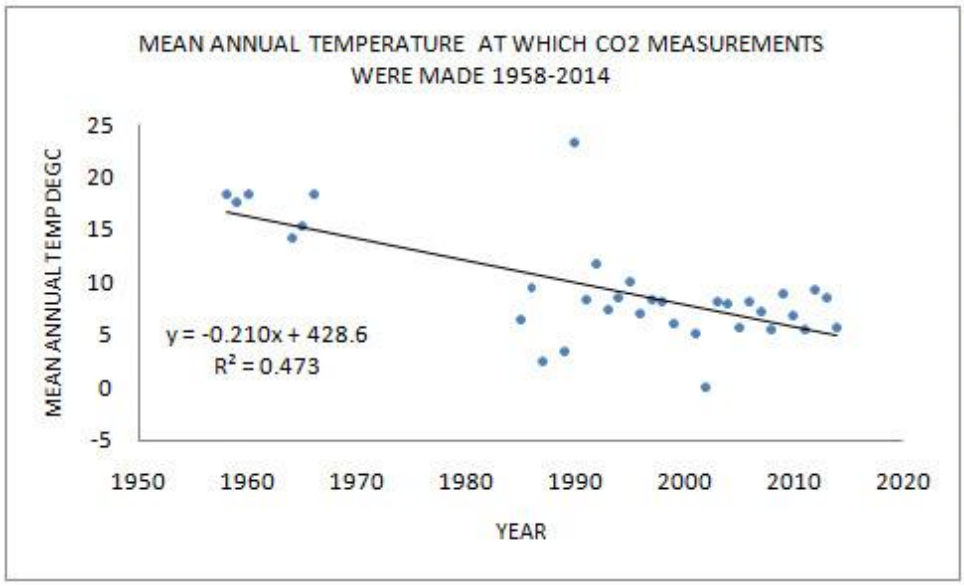
-0.00027	0.00
0.01830	0.00735
0.00001	0.03822
t-stat=	0.02
p-val=	0.98815

3.2 Case 2: Annual means with temperature correction, equally weighted, no uncertainty

Figure 7 shows that the CO2 concentrations presented in Section 3.1 may be biased because the temperatures at which the measurements were made are neither uniform nor random across the sample period. The measurement temperature shows a statistically significant declining trend. Since the Henry's Law equilibrium constant K_H for CO2 is sensitive to temperature (Sander, 2015) it is necessary to check how the temperature bias affected the results in Section 3.1 by converting all CO2 measurements to their equivalence at 25°C.

The temperature-adjusted CO2 concentrations appear in Figure 8 and they show that the effective CO2 concentration increased much faster than the rate indicated in Section 3.1 where the CO2 time series was biased by lower measurement temperatures in later years. It is therefore necessary to examine the data in Figure 8 to determine whether a year to year relationship exists between fossil fuel emissions and changes in the effective oceanic CO2 concentration.

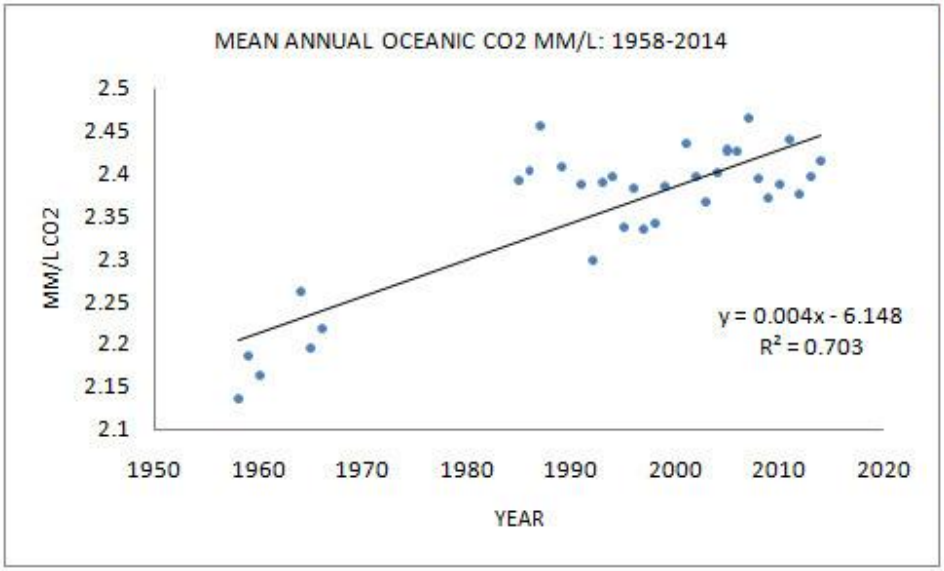
Figure 7: Mean annual temperature at which CO2 measurements were made



REGRESSION RESUTS

-0.2104241	428.661403
0.0392118	78.159823
0.4736644	3.774031
tstat=	5.366350
pval=	0.000006

Figure 8: Trend in temperature adjusted CO2 values



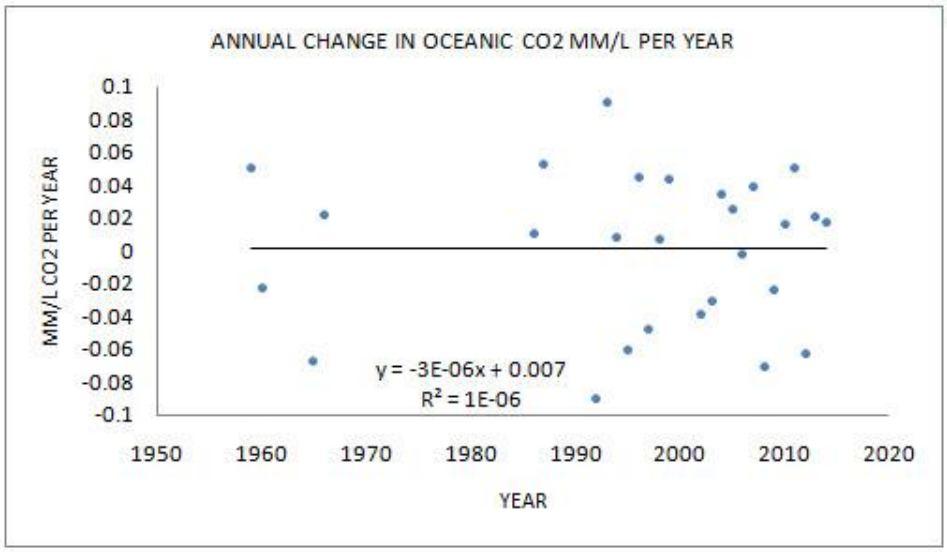
REGRESSION RESULTS

0.004267	-6.148842
0.000498	0.992811
0.703025	0.047909
tstat=	8.566557
pval=	0.000000

The year to year changes in temperature adjusted CO2 are shown in Figure 9 and the detrended CO2 series is compared with detrended fossil fuel emissions in Figure 10. The correlation between the detrended series is depicted in Figure 11. As in Section 3.1, we find no evidence that annual changes in oceanic CO2 are related to the annual rate of fossil fuel emissions. This relationship is a necessary pre-condition for the hypothesis that fossil fuel emissions cause ocean acidification.

All data and computational details for the results presented here are available in the data archive for this paper (Munshi, Acidification Paper Archive, 2015).

Figure 9: Annual changes in temperature adjusted CO2



REGRESSION RESULTS

-3.1076E-06	0.00713
5.7395E-04	1.14562
1.1727E-06	0.04735
tstat=	0.00541
pval=	0.99572

Figure 10: Detrended emissions and detrended changes in oceanic CO2

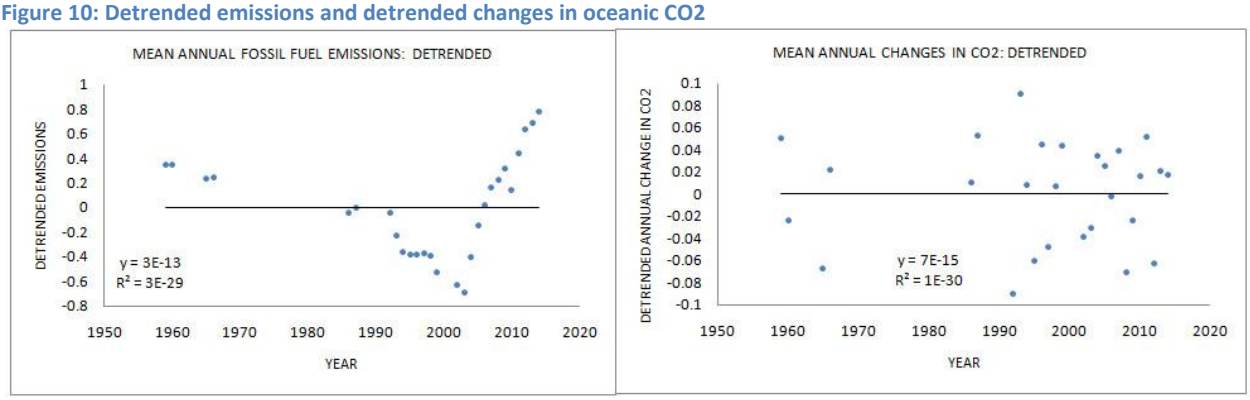
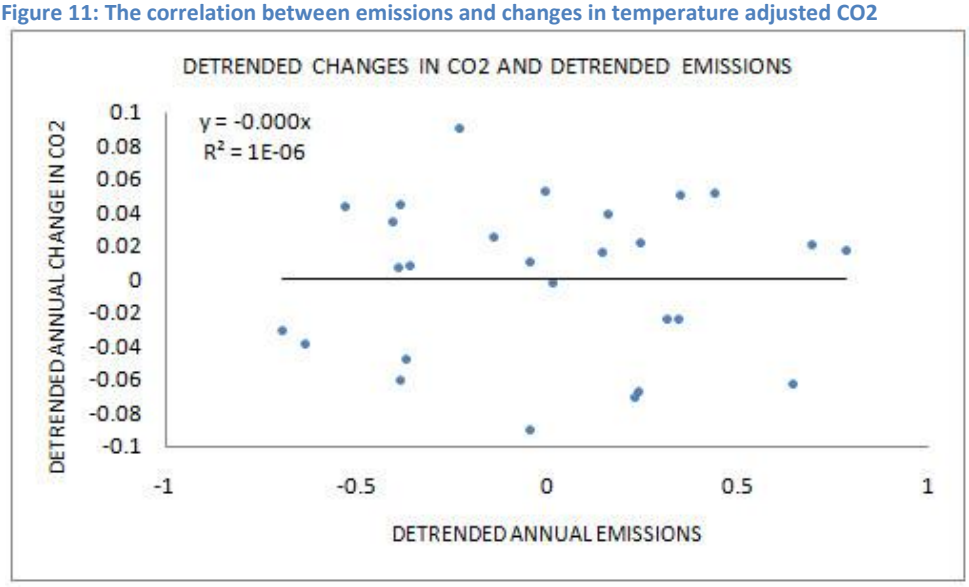


Figure 11: The correlation between emissions and changes in temperature adjusted CO2



REGRESSION RESULTS

-0.00012	2.99E-18
0.022667	0.009112
1.03E-06	0.047347
tstat=	0.005081
pval=	0.995988

3.3 Case 3: Annual means with temperature correction, equally weighted, bootstrap uncertainty

The mean and standard deviation information for the CO2 and emissions data are used to create a synthetic dataset using a Monte Carlo bootstrap procedure. The synthetic dataset is structured in a way that allows annual differences in oceanic CO2 to be computed.

The original CO2 dataset contains 124,813 observations but only 77,778 annual differences can be computed because the number of differences is limited by the smaller of the two sample sizes. The average number of differences per year is 2,880. For the equally weighted synthetic dataset we generate 2,880 simulated values of emissions and CO2 differences per year and for each of the years in which a difference can be computed. We then apply detrended analysis to the large simulated sample of 77,600 CO2 change and emission values. The procedure is demonstrated in Figures 12 and 13.

Figure 12: Monte Carlo procedure for generating the synthetic sample

	A	B	C	D	E	F	G	H
1	from	to	DCO2	stdev	co2sim	EMIS	stdev	em-sim
2	2012	2013	0.0210	0.2455	0.0383	9.68	0.2420	9.4669
3	1964	1965	-0.0664	0.2552	-0.1918	2.995	0.0749	2.9161
4	1997	1998	0.0071	0.1545	-0.1645	6.651	0.1663	6.5028
5	1996	1997	-0.0476	0.2045	0.0791	6.542	0.1636	6.2380
6	1998	1999	0.0441	0.1744	0.1464	6.643	0.1661	6.6851
7	1959	1960	-0.0229	0.2018	-0.2039	2.454	0.0614	2.5553
8	1965	1966	0.0225	0.2600	-0.1545	3.13	0.0783	3.0425
9	2003	2004	0.0352	0.2348	-0.0318	7.416	0.1854	7.6172
10	1996	1997	-0.0476	0.2045	-0.2421	6.542	0.1636	6.3520
11	2003	2004	0.0352	0.2348	-0.0659	7.416	0.1854	7.3155
12	2009	2010	0.0164	0.2217	0.5796	8.74	0.2185	8.4149
13	2012	2013	0.0210	0.2455	-0.1157	9.68	0.2420	9.6087
14	1964	1965	-0.0664	0.2552	-0.6112	2.995	0.0749	2.9092
15	2008	2009	-0.0239	0.2310	-0.3355	8.783	0.2196	8.7544
16	1995	1996	0.0457	0.2350	0.1780	6.398	0.1600	6.5161
17	2008	2009	-0.0239	0.2310	-0.1874	8.783	0.2196	9.1551
18	2009	2010	0.0164	0.2217	0.0615	8.74	0.2185	8.7014

For each annual difference year-pair (columns A and B) for which data are available, a random sample of 2,880 values are taken from the distribution of annual differences in CO2 described by the mean and standard deviation of the observed differences (columns C and D) and these simulated differences are placed in column E. These values correspond with the "to" year in column B. Similarly, a random sample of 2,880 values are taken for each "from" year in column A using the mean and standard deviation in columns F and G. The result is placed in column H. We can now use linear regressions of column E against B and column F against A to generate the detrended series in columns J and L in Figure 13.

Column P in Figure 13 contains the correlation between the detrended series. The high p-value indicates that the data do not provide evidence of a relationship between annual changes in oceanic CO2

concentration and annual fossil fuel emissions. All data and computational details are available in the data archive for this paper (Munshi, Acidification Paper Archive, 2015).

Figure 13: Detrended correlation analysis of the simulated data

	I	J	K	L	N	O	P	Q
1	dco2.hat	dc02-det	emis-hat	emis.det	detrend dco2			
2	0.0002	0.0380	8.8562	0.6107	-2.187E-05	4.425E-02		
3	0.0013	-0.1930	2.6284	0.2876	5.226E-05	1.043E-01		
4	0.0006	-0.1651	6.9100	-0.4073	2.251E-06	2.314E-01		
5	0.0006	0.0785	6.7803	-0.5422	detrend emis			
6	0.0005	0.1459	7.0398	-0.3547	0.1297	-252.1906		
7	0.0014	-0.2053	1.9797	0.5756	0.0001	0.1982		
8	0.0013	-0.1558	2.7582	0.2843	0.9564	0.4396		
9	0.0004	-0.0322	7.6885	-0.0713				
10	0.0006	-0.2427	6.7803	-0.4282		Correlation analysis		
11	0.0004	-0.0663	7.6885	-0.3730		With trend	Detrended	
12	0.0003	0.5793	8.4670	-0.0521	r	-0.0023	-0.0038	
13	0.0002	-0.1159	8.8562	0.7525	n	77600	77600	
14	0.0013	-0.6125	2.6284	0.2808	sqrt(n)	278.6	278.6	
15	0.0003	-0.3359	8.3372	0.4172	r2	0.00001	0.00001	
16	0.0006	0.1774	6.6505	-0.1345	1-r2	0.99999	0.99999	
17	0.0003	-0.1877	8.3372	0.8179	stdev	0.00359	0.00359	
18	0.0003	0.0612	8.4670	0.2344	t	0.63093	1.06432	
19	0.0003	0.1577	8.4670	0.5669	p	0.52809	0.28719	

3.4 Case 4: Monte Carlo simulation of decadal moving averages

In this Monte Carlo simulation we use the regression equation in Figure 9 to estimate values for oceanic CO2 concentration for all years in the sample period. Three thousand simulated samples are taken from the distribution implied by Figure 9. The uncertainty implied by the standard error of the regression coefficients is included in the simulation. We use these simulated samples to check whether a 10-year moving average smoothing of the random variability in the changes in oceanic CO2 will reveal a correlation between annual fossil fuel emissions and annual changes in oceanic CO2 concentration.

Six of the samples are depicted graphically in the figures used in this section. The left panel of Figure 14 shows the predicted values of CO2 concentration according to the regression model in Figure 9 for each year in the period 1958-2014 including the years of missing data. The panel on the right shows their moving averages for the period 1967-2014. The annual differences between the moving averages appear in the right panel of Figure 15.

The detrended series are shown in Figure 16. Three thousand correlations are computed between them one for each simulated sample. Their average is r = -0.00269 and their standard deviation is σ = 0.1463. The correlation is not statistically different from zero. We fail to reject H_0 and thus find no evidence in the moving average experiment of a relationship between emissions and the CO2 concentration of the oceans.

Figure 14: Simulated values of CO2 concentration 1958-2014 and their 10-year moving averages 1967-2014

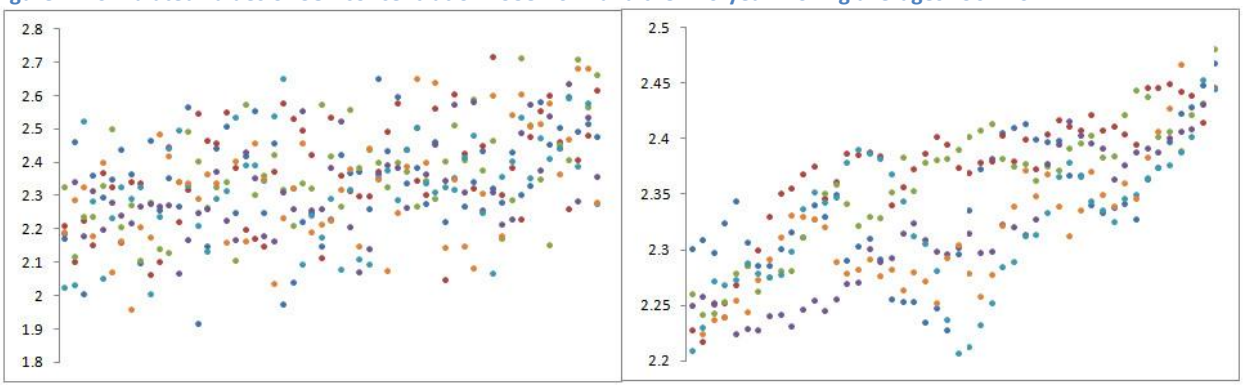


Figure 15: Annual fossil fuel emissions and annual changes in the 10-year moving average of CO2 concentration

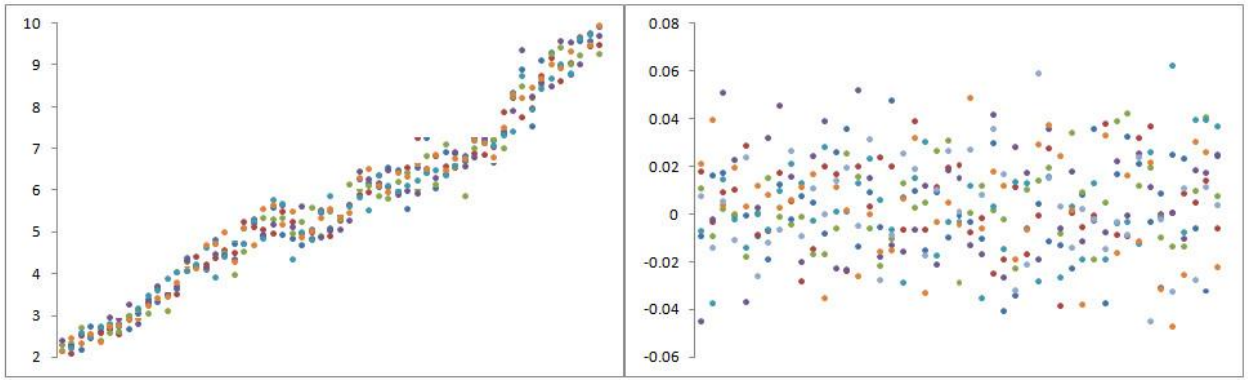
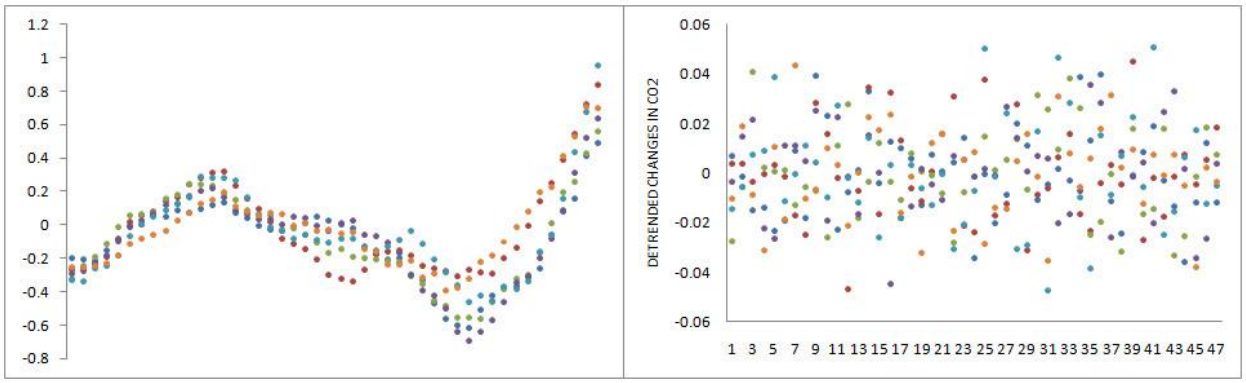


Figure 16: The two series detrended



All data and computational details are available in the online data archive for this paper (Munshi, Acidification Paper Archive, 2015).

4. CONCLUSION

Figure 2, Figure 8, and Figure 14 leave no doubt that the data in the World Ocean Database of the NOAA show a rising trend in oceanic CO2 concentration for the period 1958-2014 and it is possible that these changes may have consequences for marine life and fisheries as many laboratory experiments (Cornwall, 2015) and climate model experiments (Caldeira K. , 2003) (McNeil, 2006) have suggested. However, it is not clear from the data that these effects can be mitigated in any way by reducing fossil fuel emissions.

In this empirical study of historical emissions data and historical CO2 concentration data of the oceans over a 57-year period from 1958 to 2014, we were unable to detect a correlation between the annual rate of emissions and the mean annual change in oceanic CO2. This correlation is a pre-condition to the anthropogenic ocean acidification hypothesis which holds that the annual rate of human emissions causes annual changes in oceanic CO2 concentration (Scripps, 2013) (NOAA-1, 2015). It is therefore suggested that the study of these changes should broaden the scope of the investigation beyond anthropogenic effects to include long term natural variability.

These findings are presented with the disclaimer that the CO2 time series used in the study contains gaps that constitute more than one third of the span in the study period. All data and computational details used in this paper are available in an online data archive (Munshi, Acidification Paper Archive, 2015).

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