HAS THE AMOUNT OF CARBON DIOXIDE IN THE ATMOSPHERE CHANGED SIGNIFICANTLY SINCE THE BEGINNING OF THE TWENTIETH CENTURY?

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ABSTRACT

The search for causes of the rising temperatures in some geographic areas during the twentieth century has directed interest toward the amount of atmospheric carbon dioxide (CO_2). If the carbon dioxide added by the combustion of fossil fuels remains as a net increase, any temperature-changing effects of its presence as a minor constituent of the atmosphere should be cumulatively operative as the amount increases.

In this paper, the physical knowledge of atmospheric CO_2 is examined and the available nineteenth and twentieth century observations of the atmospheric CO_2 concentration are summarized to ascertain the extent to which they corroborate claims that the amount of atmospheric CO_2 has increased since the nineteenth century. In the light of the uncertainty of both physical knowledge and of statistical analysis, it is concluded that the question of a trend in atmospheric CO_2 concentration remains an open subject.

1. INTRODUCTION

This report examines the physical knowledge of atmospheric CO_2 and summarizes the available nineteenth and twentieth century observations of the atmospheric content of CO_2, to ascertain how far they corroborate claims that the amount of carbon dioxide in the atmosphere has increased since the nineteenth century. Charts and tables are included, showing the locations, periods of record, numbers of measurements, and the ranges of values of the observed CO_2 concentration.

The amount of carbon dioxide in the atmosphere is actually little more than a trace [18, 23]. That dissolved in the waters, or combined as carbonates, etc., in the crust of the earth, is much greater. Goldschmidt [15] presents a table, showing the location of the earth's CO_2 and potential CO_2. It is the source for table 1. From this table, it appears that about 0.005 percent of the earth's crustal carbon is in the atmosphere as CO_2.

Important climatic effects are attributed to this small percentage of carbon dioxide in the air, and, according to Callendar [6, 7, 9] and Plass [24], a significant increase in the concentration of CO_2 would noticeably raise the surface temperature of the earth because of the "greenhouse effect."

TABLE 1.—*Kilograms of CO_2 and potential CO_2 per square centimeter of the earth's surface (After Goldschmidt [15])*

A. In the carbonates in sedimentary rocks	6.56.
B. In coal, bitumen, humus, and the biosphere	0.67 to 3.1.
C. In sea water	0.020.
D. In the atmosphere, mainly CO_2	0.0004.
Total	7.3 to 9.7.

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In 1938, Callendar [6] suggested that the combustion of the fossil fuels, such as coal, lignite, petroleum, etc. may be causing such an increase. At that time, according to his estimate, 4.3×10^9 tons of CO_2 per annum were being added to the atmosphere in this way. He gave the total added between 1887 and 1937, after allowing for an accelerated rate of burning as time went on, as about 1.5×10^{11} tons. So large an amount added so quickly would, he suggested, be absorbed into the earth's waters at a much slower rate. Assuming that other natural processes, such as the biological exchange, be in balance, the result would be an increase of atmospheric CO_2 with time. He estimates that 2000 to 5000 years will be required before we may expect the atmospheric content to reach equilibrium with the rate of oceanic absorption.[1] Since the acceleration in the rate of industrial combustion may not cease for some time [17], the consequently increasing CO_2 in the atmosphere would increase the absorption of outgoing radiation, and the surface layer of the atmosphere would become warmer.

In support of this view, Callendar [7] selected from the published records of determinations of CO_2, made between 1867 and 1935, those which he considered the most accurate. On the evidence of these records, he found that the CO_2 had apparently increased since 1900 by about 6 percent.

Figure 1, after Glueckauf [14], shows the increase in content, according to Callendar's selected data. To Glueckauf's plotted points have been added more recent data, including Callendar's 1949 computed value [9], and other

[1] Suess [26], however, estimates that the "average lifetime of a CO_2 molecule in the atmosphere before it is dissolved in the sea will lie between 20 and 50 years."

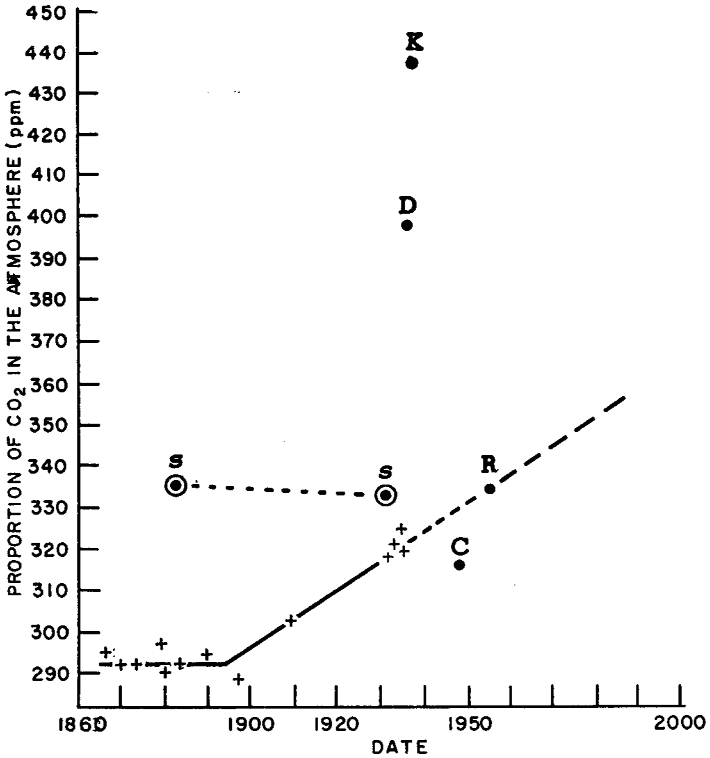


FIGURE 1.—Trend in CO_2 content of the earth's atmosphere (selected data). Adapted from Glueckauf [15]. To Glueckauf's values (+) have been added those of Duerst (D), 500 observations [13]; Kreutz (K) 25,000 observations [21]; Callendar's latest estimate, 1949 (C) [9]; and the means derived in the present paper (S).

recent estimates mentioned at various points in the present paper.

Buch [5], used his own observations (1932–1935) taken in scattered high latitudes in the North Atlantic ocean and its estuaries, as representative of the concentration of atmospheric carbon dioxide at that time, and the same sources as those selected by Callendar to represent the content in the late nineteenth and early twentieth centuries. Then, comparing these latter data with his own newly observed values, he came to substantially the same conclusion as did Callendar.

Recently there have been independent studies which are at least consistent with Callendar's. Among these recent studies may be mentioned those of Brown [3], who, in 1952, determined the C^{12}/C^{13} ratio in tree ring samples. He found evidence that this ratio is, on the average, greater in the younger samples than in the older. This indicates, he suggests, that "carbon dioxide in the air has been diluted in recent years by carbon dioxide from industrial sources," and [4] that the total "Carbon dioxide content of the atmosphere may be increasing, or at least may not be in equilibrium with the oceans." Dingle [11], by physical reasoning, arrives at the conclusion that the CO_2 content of the atmosphere at present exceeds 0.03 percent, which is in excess of the proportion Callendar estimates for the nineteenth century.

Hutchinson [19] has stated that, "There can be little doubt that during the first half of the twentieth century the mean CO_2 content of the air in north temperate latitudes has increased." Callendar has thus had a number of supporters in whole or in part.

Independent opinion has not, however, been unanimous in support. At the time Callendar delivered his 1938 paper [6], Mr. J. H. Coste suggested that the accepted CO_2 content had at the turn of the century been considered to be about 0.04 percent, and not the 0.029 percent indicated by the measurements Callendar cited. Mr. Coste then asked, since the value, 0.04 percent, is a higher percentage than the average value of about 0.032 percent Callendar found for the 1937 CO_2 content, can we be sure that there has been any net increase at all in the percentage of CO_2 in the atmosphere?

2. PHYSICAL EVIDENCE

There are processes which may deplete the increased concentration of CO_2 produced by combustion, and others which may be more important than combustion of fossil fuel in increasing the concentration, at least temporarily. Callendar does not consider them relatively important, but it seems logically tenable to suppose that a relatively slight increase in the rate of biologic absorption of CO_2 might nearly, or even more than, compensate for any increase in its evolution from other sources, such as industrial combustion.

Moreover, Hutchinson [19] suggests that with the expansion of industry through the past century, agriculture also expanded, and that there is a far greater opportunity for loss of respiratory CO_2 from soil in arable land than in forest land. He therefore doubts the validity of Callendar's explanation of the source of an increase in the amount of CO_2.

Dingle [11], in discussing Callendar's findings, points out the complexities of determining the amount of CO_2 in the world's atmosphere as a whole. Measurements at one or a few localities over a limited period of time are inadequate, since the concentration of carbon dioxide varies in air masses of differing trajectories. He suggests that increases in the concentration of observed CO_2 might be due to changes in the general atmospheric circulation rather than necessarily mainly due to a worldwide increase in CO_2 concentration. He holds this to be a more attractive physical hypothesis to explain any increase in the observed CO_2 value than is Callendar's thesis that the higher temperatures are due to an increasing concentration in the atmosphere as a whole.

Since this paper was initially prepared for publication, two studies by Suess [25, 26] have become available. He cites the fact that fossil carbon does not contain appreciable C^{14}, and presents evidence that the proportion of C^{14} contained in tree rings has decreased slightly since the nineteenth century. In its place is a greater proportion of C^{12}. The decrease in the ratio is, however, greatest

Table 2.—*Physical and chemical processes adding or extracting massive quantities of CO_2 to or from the atmosphere (after Hutchinson [19])*

Process or category	Gross emission ($\times 10^{12}$ grams/year)		Gross absorption ($\times 10^{12}$ grams/year)	
	At least	But not more than	At least	But not more than
Volcanic output of CO_2 [19]	30	70		
Oceanic sedimentation of Carbonate [19]			30	70
Oceanic Emission [19]	700	2, 500		
Oceanic Absorption [19]			700	2, 500
Continental Photosynthesis [2]	87, 500	88, 500		
Continental Respiration [2]			87, 500	*92, 000
Consumption of fuels	4, 000 [16]	6, 100 [20]		
Grass and forest fires [19]	5, 750			
Total Atmospheric Carbon Dioxide is probably between 2,250,000 and 2,750,000 $\times 10^{12}$ grams				

*Adding to the maximum photosynthesis estimate, a half of the highest estimate of production by industrial consumption of fuels.

near concentrations of industry, and is much less in the case of a tree which grew in Alaska than in the cases of trees near dense population centers. Suess concludes that ". . . the world-wide contamination of the earth's atmosphere with artificial CO_2 probably amounts to less than one percent."

Brown's [3, 4] evidence from tree rings of the dilution of CO_2 has been suggested as showing that there has been an increase in the amount of CO_2. This does not necessarily follow. It can be shown that dilution would be the expected result of any large replacement of natural by fossil carbon dioxide. Such dilution would occur whether the total amount in the atmosphere be gradually increasing, remaining approximately stationary, or decreasing. Indeed, the replacement would be most pronounced, and, therefore, most detectable in an atmosphere with decreasing CO_2 concentration. Thus far only Brown's abstracts are available. In them, he states his conclusions in strictly qualitative terms. Demonstration that the trend is either up or down awaits a quantitative discussion of his findings and of whatever compensating forces in either direction enter as complications. What Brown seems to have confirmed thus far is the already established fact that great quantities of fossil carbon have been turned into atmospheric CO_2.

Thus, students of the subject differ. That is, physical reasoning has not as yet shown that CO_2 is necessarily increasing as a result of the addition of combustion gases. There remains the statistical approach, that used by Callendar. The current knowledge from a quantitative standpoint is summarized in table 2.

3. STATISTICAL EVIDENCE

With a dependence on statistical evidence, the mathematically established statistical criteria for significance of results must rule the degree of confidence with which conclusions may be drawn from the original data. Callendar's and Buch's averages appear, as presented in figure 1, to show an increase in CO_2 from the late nineteenth century to the beginning of the middle third of the twentieth. Their comparisons are, however, based on a narrow selection of values from a much larger body of data, scattered through the scientific literature of the past century. It may be granted that the data they used are probably quite accurate averages for the time, place, meteorological conditions, etc., of observation. The question remains, however, are all the measurements which they did not use, inaccurate?

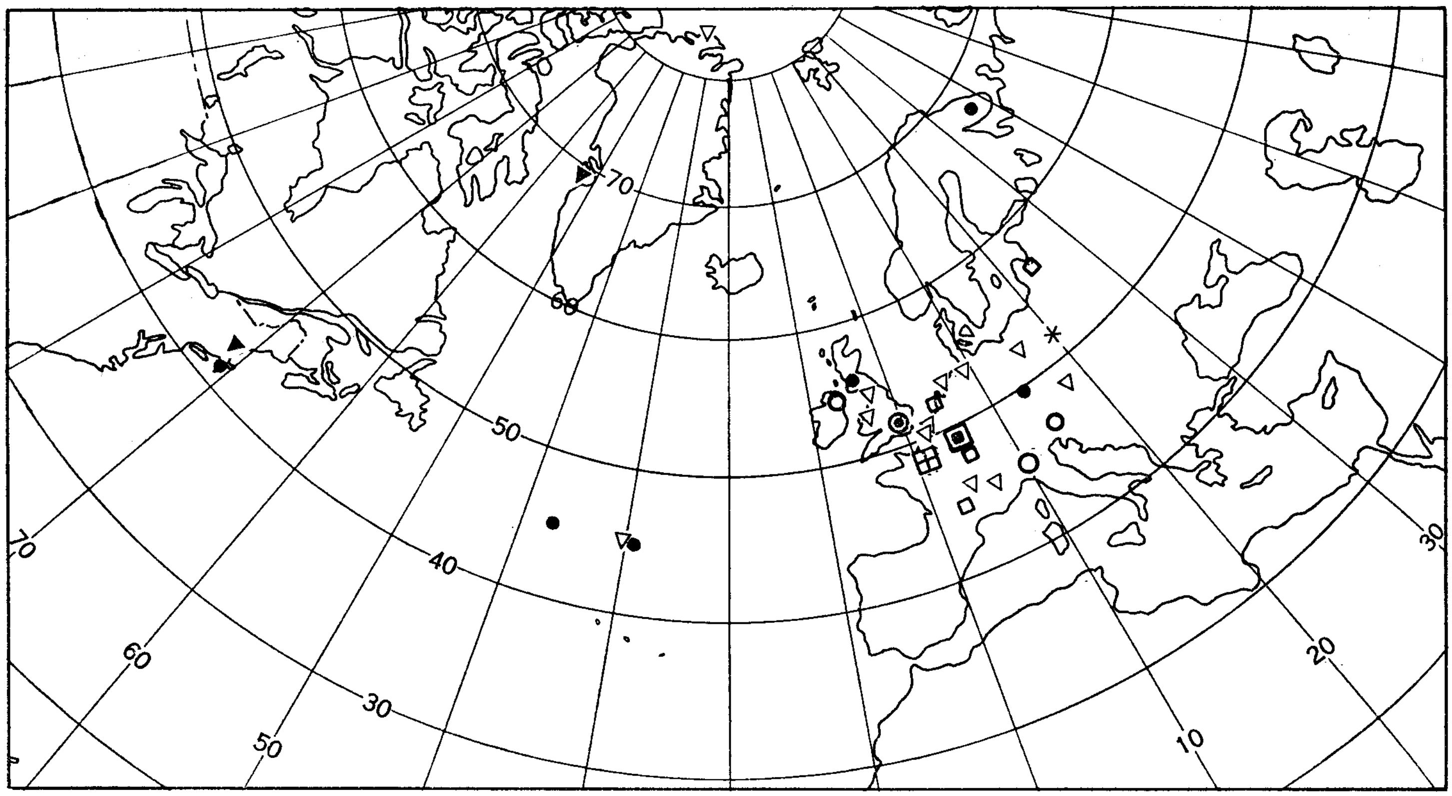
Buch who followed Callendar, accepts without challenge Callendar's selection of data, and merely adds his own observational material. Hutchinson [19] bases his statement that CO_2 has increased on Callendar's results, although he limits his corroboration to the case of the air in the North Temperate zone. Brown and Dingle alone offer any new evidence on the amount of CO_2 in the atmosphere, and their evidence, if it be not negative, is not necessarily confirmatory.

Since Callendar, by basing his hypotheses on statistical data, has tacitly invoked the laws of statistical evidence, it is fitting to examine the validity of his procedure, that of using only the data he believed to be of the best quality available, rejecting the rest.

The mathematics of statistics, and the experience of statisticians both indicate, as a general principle, that arbitrary rejection of data, without specific knowledge of their unreliability or unapplicability, is questionable. Although the purpose of such a procedure may be to remove an observational or sampling bias that is known to be present, selection of the data to be used will often introduce a greater source of error than that which it was intended to remove.

At best, the omission of part of the data is not as necessary or as helpful as might appear at first thought, since it can be shown that when the means of two sets of data are compared, the presence of a given average bias in each set will not affect the difference nor the standard error of this difference, except as an added contribution to the variance of the sample. If, however, some of the data be selected to the exclusion of the rest, for the purpose, perhaps, of reducing the magnitude of the residual variance, due to crudity in some of the measurements, then, in addition to any unintentional bias that might be introduced in the comparison of the means, there might also result an underestimate of the standard error of the difference, due to the mistaken rejection of those of the extreme values which actually belong to the distribution. The result may be an entirely spurious accuracy in the means, which leads to unjustified conclusions.

In the light of these considerations, a reexamination of the entire body of available measurements of the relative proportion of CO_2 in the atmosphere may have some value. Fortunately, Effenberger [13] has compiled what seems to be a fairly complete list of the published observations up to 1940. He has indicated the sets of determinations used by Callendar [8]. More recently, the American Meteoro-



Symbol	Period of Observations	Location	No. of Obs.	CO_2 content of atmosphere (parts per million) Min.	Mean	Max.
⊞	1816-1827	France	- -	370	410	620
*	1844	Prussia	- -	210	400	420
▽	1866-1879	45°N, 30°W	4	- -	270	- -
▽	1866-1879	Greenland	3	480	550	640
▽	1866-1879	England	26	- -	310	- -
▽	1866-1879	England	53	210	296	410
▽	1866-1879	France	80	270	292	350
▽	1866-1879	France	89	- -	291	- -
▽	1866-1879	Germany	1,034	270	292	350
▽	1866-1879	Germany	347	210	330	420
▽	1866-1879	Austria	295	300	340	410
▽	1866-1879	Switzerland	- -	210	330	420
▽	1866-1879	France	- -	- -	410	- -
▽	1866-1879	France	- -	- -	300	- -
□	1880-1889	Belgium	525	260	294	350
□	1880-1889	E. Baltic	266	- -	300	350
□	1880-1889	France	1,000	240	292	360
□	1880-1889	France	64	- -	287	- -
□	1880-1889	France	- -	- -	290	- -
○	1890-1898	Ireland	64	- -	289	- -
○	1890-1898	England	92	- -	294	- -
○	1890-1898	Austria	- -	200	380	550
▼	1904-1919	U.S.	645	- -	303	- -
▼	1904-1919	Greenland	59	- -	480	700
■	1920-1929	France	17 137	180	290 310	590
●	1930-1939	U.S.	- -	- -	329	- -
●	1930-1939	45°N, 37°W	28	152	318	368
●	1930-1939	45°N, 29°W	53	- -	320	- -
●	1930-1939	Scotland	152	- -	324	- -
●	1930-1939	England	- -	- -	310	350
●	1930-1939	Finland	95	- -	321	- -
●	1930-1939	Germany	25,000	- -	438.5	- -
●	1930-1939	Italy	500	240	400	790

FIGURE 2.—Geographic distribution of selected CO_2 content measurements that have been made in the Northern Hemisphere and the data available for each location.

logical Society [1] has published a "Bibliography on Carbon Dioxide in the Atmosphere." From this source and elsewhere, references have been found and some additional, more recent, data have been compiled. The geographic distribution of these observations of CO_2 atmospheric content and other data listed by Effenberger and the other sources used in these summary tables, are shown in figure 2. This figure shows the means and the highest and the lowest values of the atmospheric concentration found during each of the observational programs represented. Where available, the numbers of observations, on which the means were based, are given.

The asterisk and boxed plus sign in figure 2 show the data for the observations which were made earlier than the first of those selected by Callendar. One set of observations was made in 1816, the other in 1844. The observations shown by open symbols represent the period 1866–1901 from which Callendar selected his values for the latter part of the nineteenth century. During this period, the consumption of fossil fuel had not become as great as it was between 1901 and 1930, a period represented roughly by the bulk of the data charted as solid symbols. Where a closed symbol appears inside an open symbol, observations were made during both of the latter periods.

In table 3, the mean values shown in figure 2 are reclassified to show the values used by Callendar for the

TABLE 3.—*Mean CO_2 values, in parts per million. Determined by observation in the period 1816 to 1940. Compares CO_2 content for observations used by Callendar with that for observations not used by him. [Arrangement is in order of magnitude.] (after Effenberger [13] except as noted)*

Nineteenth Century (1816 to 1901)			Twentieth Century (1904 to 1935) (1936 to 1940) 1904			
				Means not used by Callendar		
Means used by Callendar	Means not used by Callendar	Documented as anomalous data	Means used by Callendar	Rejected as inaccurate etc.	Published too late to be available	Documented anomalous data [22]
287	270	[a] 460	303	290	400	[c] 480
289	290	[b] 560	318	310	438.5	
291	300		320	[d] 310		
292	300		321			
292	320		324			
292	330					
294	330					
294	340					
295	350					
296	380					
	400					
	410					
	410					
	470					
	550					
Means 292	363		317	303	419	
Combined means 335			334			

[a] Observed on rainy days
[b] Observed on days with snow
[c] Krogh's [22] data
[d] Not listed by Effenberger. Cited by Callendar, but not included in his means

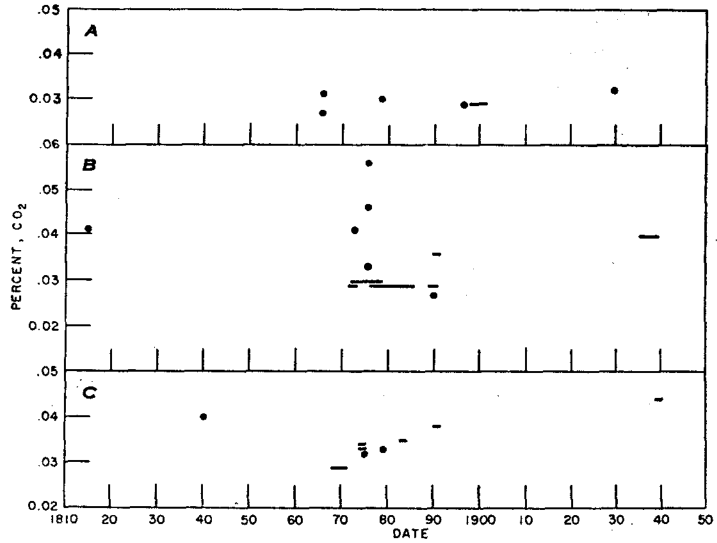


FIGURE 3.—Proportional amounts of atmospheric CO_2, in parts per million, measured in (A) the British Isles, (B) France and Switzerland, and (C) Germany, Denmark, East Baltic States, and Austria-Hungary. Length of line denotes length of time observational program continued. Dots are used for periods less than one year. The line segment in (C), at 1939–40 and showing 438.5 ppm, is based on more observations than all other points and line segments on all three charts combined.

nineteenth and for the twentieth centuries, and three categories of observations not used by him. Each value, as in figures 1 and 2, is a mean of a group of observations, varying from 3 to about 25,000.

Figure 3 shows the majority of these determinations grouped in another way. Here, the means of the sets of observations, for each of the principal regions where measurements were made, are shown for: British Isles (fig. 3A); France and Switzerland (fig. 3B); Central Europe, including Germany, Austria-Hungary, the eastern Baltic States, and Denmark (fig. 3C). The length of line, representing each mean, shows the length of time the observational program continued. It can be seen from this figure that the majority of programs were of short duration, and from the table accompanying figure 2 that in some cases only a few observations were made.

Reference to the three charts in figure 3 does not reveal any significant trend in CO_2 content, such as is so clearly shown in figure 1. Indeed, after excluding values which the observers themselves have designated as non-representative, but not any of the others, then the mean value for the nineteenth century is 335, and for the first third of the twentieth century 334 parts per million. Such a close approach to identity of values for the two periods is, of course, an accident. Referring to the texts of the papers from which Effenberger made his tabulations, it appears that there has been wide variability in the means found for differing geographical regions, on land and on sea, and from one synoptic weather condition to another. The data-gathering programs were conducted by mutually independent observers, using differing techniques. There are so many possible sources of variability, that there is no basis for any claim, based on these data, that the CO_2 content of the atmosphere has remained anywhere near constant. Similarly, there is inadequate basis for a claim that Glueckauf's trend line approximates the recent trend of the actual carbon dioxide content of the earth as a whole.

The means that Callendar rejected from the nineteenth century records are, in the main, indicative of higher values than those he accepted. He points out that the accuracy of observations improved as time went on, and that early techniques tended to give too high values. Statistically speaking, the data in table 3 could well be drawn from a population having these properties.

The three values for the twentieth century, however, which Callendar rejected average lower than those he accepted. This does not demonstrate that his choice was bad, but the fact that he considers so many nineteenth century values to be overestimates and two twentieth century values to be underestimates raises a question about his method of selection.

Since techniques have been improving, the latest observations should be the most accurate. Duerst [12] and Kreutz [21] found values of 400 and 438.5 parts per million, respectively, from observations made in 1936 and 1939. Duerst bases his mean on 500 observations, a reasonably large number, if his techniques are correct. Kreutz made about 25,000 observations. This is more than were made in all other herein listed observing programs combined. He expresses confidence in the accuracy of his measurements and of his computed mean values.

Admittedly Duerst's and Kreutz' values may be more representative of the atmospheric concentration of CO_2 at the time and place of observation than of the earth, or even the Northern Hemisphere, as a whole. By the same token, however, might not some of the high nineteenth century values and low twentieth century values be as representative as those Callendar accepted?

Callendar's presentation of his 1938 paper on the subject of increasing CO_2 occurred just after a succession of five warm years in western Europe. Since then, this positive anomaly has been persistent in some densely populated districts in the United States and western Europe; on the other hand, temperatures have been lower, rather than higher, in recent decades, than they were in the nineteenth century in some Southern Hemisphere regions [10], and elsewhere. Can we be entirely sure that the earth as a whole has warmed up enough to require an increase in CO_2 in the air to explain it?

At any rate, it is apparent that, if we use the statistical approach, different degrees of selectivity in determining which data to include are productive of differing final results.

4. CONCLUSIONS

Is the CO_2 increasing? Much seems to depend on the objectivity of Callendar's decisions as to which data to keep.

In the light of the uncertainty of both physical knowledge and of statistical analysis in determining whether the relative proportion of carbon dioxide in the air is increasing significantly, remaining almost constant, or even decreasing slightly, the final word cannot as yet be considered to have been said. Instead, the subject remains open, either until another chemist critically evaluates the accuracy of the existing data, or else until more and better-organized data are available.

All this does not refute Callendar's thesis. The available data merely fail to confirm it. The positive evidence that the CO_2 has increased is inconclusive, but seems strong enough to reward further study, and the time seems ripe for new research.

It may be hoped that the collection of standardized measurements of CO_2 can be made a part of the 1957–58 International Geophysical Year program. Once a dependable set of observational data has been assembled, the evidence of the old observations can perhaps be reevaluated. If such new reevaluation proves impracticable, even then a reliable set of new worldwide observations can serve as a basis for comparison in future years.

In summary, the data, at present available, are inadequate as they now stand to prove or disprove a statistically significant trend in CO_2 concentration in the atmosphere. If and when an upward trend has been demonstrated, and its cause ascertained, it will then be valid to base physical explanations of atmospheric events on the assumption that CO_2 is increasing. Meanwhile, Callendar's interesting extrapolations (through the 22d century) of the effects of burning up of the world's fuel, stimulate the interest of the speculatively minded.

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