RESTORING THE QUALITY OF OUR ENVIRONMENT



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President's Science Advisory Committee

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RESTORING THE QUALITY OF OUR ENVIRONMENT

an adequate number to start with. They should be selected by use nodern probability methods.

The waters of a single drainage area may vary from small trickles to ge lakes. In sampling these waters, however, we can take advantage the way in which the waters flow from smaller streams into larger s, and from larger ones into rivers and lakes.

The lands of a drainage area may vary even more widely: highly culted farm lands may grade through rangelands to rocky mountains and erts. All are part of the general environment; the pollution of all st be considered in the overall result. We can take advantage of what know about the likelihood of pollution of different kinds of land in nning the details of the sampling, but we must give all lands their propriate chance to contribute to the final result.

APPENDIX Y4

Atmospheric Carbon Dioxide

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Section I. CARBON DIOXIDE FROM FOSSIL FUELS—THE INVISIBLE POLLUTANT

INTRODUCTION

Only about one two-thousandth of the atmosphere and one tenthousandth of the ocean are carbon dioxide. Yet to living creatures, these small fractions are of vital importance. Carbon is the basic building block of organic compounds, and land plants obtain all of their carbon from atmospheric carbon dioxide. Marine plants obtain carbon from the dissolved carbon dioxide in sea water, which depends for its concentration on an equilibrium with the carbon dioxide of the atmosphere. Marine and terrestrial animals, including man, procure, either directly or indirectly, the substance of their bodies and the energy for living from the carbon compounds made by plants.

All fuels used by man consist of carbon compounds produced by ancient or modern plants. The energy they contain was originally solar energy, transmuted through the biochemical process called photosynthesis. The carbon in every barrel of oil and every lump of coal, as well as in every block of limestone, was once present in the atmosphere as carbon dioxide.

Over the past several billion years, very large quantities of carbon dioxide have entered the atmosphere from volcanoes. The total amount was at least forty thousand times the quantity of carbon dioxide now present in the air. Most of it became combined with calcium or magnesium, freed by the weathering of silicate rocks, and was precipitated on the sea floor as limestone or dolomite. About one-fourth of the total quantity, at least ten thousand times the present atmospheric carbon dioxide, was reduced by plants to organic carbon compounds and became buried as organic matter in the sediments. A small fraction of this organic matter was transformed into the concentrated deposits we call coal, petroleum, oil shales, tar sands, or natural gas. These are the fossil fuels that power the world-wide industrial civilization of our time.

Throughout most of the half-million years of man's existence on earth, his fuels consisted of wood and other remains of plants which had grown only a few years before they were burned. The effect of this burning on the content of atmospheric carbon dioxide was negligible, because it only slightly speeded up the natural decay processes that continually recycle carbon from the biosphere to the atmosphere. During the last few centuries, however, man has begun to burn the fossil fuels that were

locked in the sedimentary rocks over five hundred million years, and this combustion is measurably increasing the atmospheric carbon dioxide.

In the geologic past, the quantity of carbon dioxide in the atmosphere was determined by the equilibrium between rates of weathering and photosynthesis, and the rate of injection of volcanic carbon dioxide. On an earthwide average, both weathering and photosynthesis must speed up when the carbon dioxide content of the air is increased, and slow down when it is diminished; consequently, over geologic time the carbon dioxide in the air must have risen when volcanic activity was high, and must have gone down when volcanoes were quiescent. On a human scale, the times involved are very long. The known amounts of limestone and organic carbon in the sediments indicate that the atmospheric carbon dioxide has been changed forty thousand times during the past four billion years, consequently the residence time of carbon in the atmosphere, relative to sedimentary rocks, must be of the order of a hundred thousand years.

The present rate of production of carbon dioxide from fossil fuel combustion is about a hundred times the average rate of release of calcium and magnesium from the weathering of silicate rocks. As long as this ratio holds, precipitation of metallic carbonates will be unable to maintain an unchanging content of carbon dioxide in the atmosphere. Within a few short centuries, we are returning to the air a significant part of the carbon that was slowly extracted by plants and buried in the sediments dùring half a billion years.

Not all of this added carbon dioxide will remain in the air. Part of it will become dissolved in the ocean, and part will be taken up by the biosphere, chiefly in trees and other terrestrial plants, and in the dead plant litter called humus. The part that remains in the atmosphere may have a significant effect on climate: carbon dioxide is nearly transparent to visible light, but it is a strong absorber and back radiator of infrared radiation, particularly in the wave lengths from 12 to 18 microns; consequently, an increase of atmospheric carbon dioxide could act, much like the glass in a greenhouse, to raise the temperature of the lower air. [Water vapor also absorbs infrared radiation, both in the range of the CO₂ band centered at 15 microns, and at wave lengths near 6.3 microns. With the average concentration of water vapor in the lower air at mid latitudes, the effect of carbon dioxide absorption is reduced to about half that which would exist in an absolutely dry atmosphere. (Möller, 1963.) Ozone, which is an important constituent of the upper air, also absorbs some infrared at wave lengths around 9.6 microns, but its principal effect on air temperature is due to its absorption of ultraviolet and visible sunlight.]

The possibility of climatic change resulting from changes in the quantity of atmospheric carbon dioxide was proposed independently by the

American geologist, T. C. Chamberlain (1899) and the Swedish chemist, S. Arrhenius (1903), at the beginning of this century. Since their time, many scientists have dealt with one or another aspect of this question, but until very recently there was little quantitative information about what has actually happened. Even today, we cannot make a useful prediction concerning the magnitude or nature of the possible climatic effects. But we are able to say a good deal more than formerly about the change in the quantity of atmospheric carbon dioxide, and about the partition of carbon dioxide from fossil fuel combustion among the atmosphere, the ocean, and the biosphere.

THE RECENT INCREASE IN ATMOSPHERIC CARBON DIOXIDE

During the five years from 1958 through 1962, 5.3 x 10¹⁶ grams of carbon dioxide were produced by the combustion of coal, lignite, petroleum and other liquid hydrocarbons, and natural gas (see Tables 1 and 2).

Table 1.—Carbon Dioxide Produced by Fossil Fuel Combustion, 1950-62 [1016 grams]

Year	Coal 1	Lignite ²	Liquid Hydro- carbons ³	Natural Gas ⁴	Total	As % of Atmos- pheric CO ₂ in 1950
1950	0. 37	0. 09	0.17	0. 04	0. 67	0. 29
1951	. 38	. 09	. 20	. 05	. 72	. 31
1952	. 38	. 09	. 21	. 05	. 73	. 31
1953	. 38	. 09	. 22	. 05	. 74	. 32
1954	. 38	. 09	. 23	. 06	. 76	. 32
1955	. 41	.10	. 25	. 06	. 82	. 35
1956	. 44	. 11	. 27	07	. 89	. 38
1957	. 45	. 13	. 29	. 07	. 94	. 40
1958	. 46	. 14	. 29	. 08	. 97	. 41
1959	. 48	.14	. 32	. 09	1.03	. 44
1960	. 50	. 14	. 34	. 10	1.08	. 46
1961	. 48	. 15	. 36	.10	1.09	. 46
1962	. 50	. 15	. 39	. 11	1.15	. 49
Total	5. 61	1.51	3. 54	. 93	11.59	4.93

Assumed carbon content, coal=75 percent.

Source: Computed from Table 2.

Table 2.—World Production of Fossil Fuels—1950–62
[Millions of Metric Tons]

Year	Coal	Lignite	Liquid Hydro- carbons ¹	Natural Gas ²	Total
1950	1, 340	530	540	155	2, 565
1951	1,375	550	620	180	2, 725
1952	1,375	550	655	200	2, 780
1953	1,380	555	690	210	2, 835
1954	1,375	550	725	220	2, 870
1955	1,500	630	790	240	3, 160
1956	1,595	665	860	260	3, 380
1957	1,625	765	905	285	3, 580
1958	1,665	825	930	305	3, 725
1959	1,730	845	995	345	3, 915
1960	1,810	875	1,075	375	4, 135
1961	1,760	900	1,140	405	4, 205
1962	1,805	905	1, 235	440	4, 38
Total	20, 335	9, 145	11,160	3, 620	44, 26

¹ Includes Petroleum and Natural Gasoline.

Source: World Energy Supplies. Statistical Papers, Series J, United Nations, New York.

This is 2.25 percent of the 2.35 x 10¹⁸ grams of carbon dioxide present in the atmosphere in 1950 (assuming an atmospheric carbon dioxide concentration of 300 ppm by volume—445 ppm by weight—and a mass of 5.2 x 10²¹ grams for the entire atmosphere.) On the average during 1958–1962, the CO₂ produced each year by fossil fuel combustion was 0.45 percent of the quantity in the atmosphere.

Beginning in 1958 and extending through 1963, two nearly continuous series of measurements of atmospheric CO_2 content were made. One of these series was taken at the U.S. Weather Bureau station near the top of Mauna Loa Mountain in Hawaii (Pales and Keeling, 1965), the other at the United States scientific station at the South Pole (Brown and Keeling, 1965). The measurements were carried out on an infrared gas spectrometer, with a relative accuracy for a single measurement of about ± 0.1 ppm. The observing stations are located near the centers of vast atmospheric mixing areas, far from uncontrollable sources of contaminants. Because of these nearly ideal locations, together with the high precision of the instruments, and the extreme care with which the samples were taken, these measurements make it possible to estimate the secular trend of atmospheric CO_2 with an accuracy greater by two

² Assumed carbon content, lignite = 45 percent.

³ Assumed carbon content, liquid hydrocarbons=86 percent.

⁴ Assumed carbon content, natural gas=70 percent. (Corresponding to a mixture by volume of 80 percent CH₄, 15 percent C₂H₆ and 5 percent N₂.)

² Assumed density of 8×10^{-4} gm cm⁻³ (1000m³=0.8 ton).

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orders of magnitude than ever before. Some fifteen thousand measurements were carried out during the five-year period.

The data show, clearly and conclusively, that from 1958 through 1963 the carbon dioxide content of the atmosphere increased by 1.36 percent. The increase from year to year was quite regular, close to the average annual value of 0.23 percent. By comparing the measured increase with the known quantity of carbon dioxide produced by fossil fuel combustion, given in Table 4, we see that almost exactly half of the fossil fuel CO₂ apparently remained in the atmosphere.

Tables 3 and 4 show that between 1860 and 1940 the amount of CO₂ produced by fossil combustion, chiefly coal, was 7.9 percent, and by 1950, 10.3 percent, of the estimated atmospheric CO₂ content in 1950. By 1959, the total CO₂ production was equal to 13.8 percent of the atmospheric CO₂. Unfortunately, the accuracy of measurements of atmospheric CO₂ before 1958 is too low to allow estimates of the secular variation, and it is therefore impossible to compute directly whether the fraction of fossil-fuel CO₂ remaining in the atmosphere throughout the past hundred years has been constant, or slowly or rapidly changing.

Table 3.—Carbon Dioxide Produced by Fossil Fuel Combustion, 1860–1959
[1016 grams]

Decade	Coal 1	Lignite ²	Liquid Hydro- carbons ³	Natural Gas ⁴	Total	As % of Atmos- pheric CO ₂ in 1950
1860-69	0, 46	0. 01			0. 47	0. 20
1870-79	0.70	. 03			. 73	. 31
1880–89	1.06	. 05	0.02		1.13	. 48
1890–99	1.49	. 08	. 04	0. 01	1.62	. 69
1900-09	2. 33	.15	. 10	. 02	2.60	1.11
1910–19	3.10	. 21	.19	. 04	3.54	1.53
1920–29	3. 26	. 31	. 48	. 07	4.12	1.75
1930-39	3.16	. 35	.74	. 12	4. 37	1.86
1940-49	3.74	. 49	1.14	. 25	5.62	2. 39
1950–59	4.11	1.07	2. 44	. 62	8. 24	3. 49
Total	23. 41	2. 75	5. 15	1.13	32. 44	13. 81

¹ Assumed carbon content, coal=75 percent.

Source: Computed from Table 4.

Table 4.—World Production of Fossil Fuels, 1860–1959
[Millions of Metric Tons]

Decade	Coal	Lignite	Liquid Hydro- carbons 1	Natural Gas ²	Total
1860–69 1870–79	1,660	85	5		1,750
1880–89	2, 560	180	15		2, 755
1890–99	3, 850	285	55	10	4, 200
	5, 405	495	140	40	6, 080
1900-09	8, 455	880	310	80	9, 725
1910–19	11, 240	1, 270	590	155	13, 255
1920–29	11,850	1,875	1,510	285	15, 520
1930–39	11,500	2, 135	2, 335	485	16, 455
1940–49	13, 570	2, 995	3, 605	970	21, 140
1950–59	14, 960	6, 465	7, 710	2, 400	31, 535
Total	85, 050	16, 665	16, 275	4, 425	122, 415

¹ Includes petroleum and natural gasoline.

Sources: From 1860 to 1949, United Nations, Department of Economic and Social Affairs: "World Energy Requirements". Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Vol. 1, pp. 3–33, 1956. For 1950–59, United Nations, World Energy Supplies. Statistical papers, Series J, United Nations, New York.

There are measurements of a somewhat different kind, which can give us useful information however. These are determinations of the socalled "Suess Effect."

PARTITION OF CARBON DIOXIDE AMONG THE ATMOSPHERE, THE OCEAN, AND THE BIOSPHERE

Because the fossil fuels have been buried for millions of years, they contain no Carbon 14. (This radioactive isotope is produced in the atmosphere by cosmic ray bombardment of nitrogen, and it has a half-life of only about six thousand years.) Consequently, the addition to the atmosphere of CO₂ from fossil fuel combustion, and its subsequent partition among the atmosphere, the ocean, and the biosphere will cause the ratio between radioactive and nonradioactive carbon to decrease. This manifests itself as a measurable reduction in the amount of radioative carbon, for example, in tree rings grown in recent years compared with rings that grew during the 19th Century. This reduction, due to fossil fuel combustion, is called the "Suess Effect," after Professor Hans Suess, who first observed it (see Revelle and Suess, 1956).

² Assumed carbon content, lignite = 45 percent.

³ Assumed carbon content, liquid hydrocarbons=86 percent.

⁴ Assumed carbon content, natural gas=70 percent. (Corresponding to a mixture by volume of 80 percent CH_4 , 15 percent C_2H_6 and 5 percent N_2).

² Assumed density of 8 x 10^{-4} gm cm⁻³ (1000m³=0.8 ton).

The measurement of the "Suess Effect" is beset with many difficulties, the chief among them being that the Carbon 14 content of the atmosphere—more precisely the ratio of C¹⁴ to C¹²+C¹³—varies by 1 or 2 percent from century to century apparently depending on the long-term variations in sunspot intensity. Tree ring measurements show that this ratio went up during the 15th and 17th Centuries, and down during the 16th and 18th, in each case by about 2 percent (Suess, 1956). Prior to the atomic weapons tests of the mid-Fifties, the ratio was lower by between 1 and 2 percent than in the middle of the 19th Century. This change during the past hundred years was apparently largely due to the "Suess Effect."

Taking the "Suess Effect" as between 1 and 2 percent over the period from 1860 to 1950, during which time the total carbon dioxide produced from fossil fuels was 10% of the atmospheric CO₂, and assuming that presently occurring changes in the magnitude of the "Suess Effect" are in the same proportion to CO₂ from fossil fuel combustion as in the past, we can compute both the relative sizes of the oceanic and biosphere reservoirs that are taking up part of the added CO₂, and the partition of CO₂ between these reservoirs. The amount taken up in the biosphere will be different if there are two or more sources of additional carbon dioxide, than if fossil fuels are the only source, but the amount absorbed by the ocean will not vary. Implied in the calculation is the further assumption that any CO₂ from other sources will have about the same ratio of C¹⁴ to total carbon as the atmosphere. Details are given in Section II.

The calculation shows that if the oceanic layer mixing with the atmosphere is several hundred meters thick, the amount of exchangable carbon in the biosphere is less than or about equal to that in the atmosphere. These are both "reasonable" values. On the other hand, if another significant source of CO2 is assumed for the last few decades, the amount of CO2 added to the biosphere becomes so large that it should have been observed. In fact, no increase in the biosphere has been noted. Perhaps the most striking result is that the ocean takes up a relatively small fraction of the total added CO₂, probably about 15%. In the past, the usual scientific belief has been that by far the larger part of any added CO2 would be absorbed in the ocean. This is undoubtedly true if we consider a sufficiently long time period, of the order of thousands or even, perhaps, hundreds of years, because the ocean as a whole contains nearly sixty times as much carbon dioxide as the atmosphere. But over shorter times, only the uppermost layers of the ocean take part in exchanges with the air. Moreover, most of the oceanic carbon dioxide is present as carbonate and bicarbonate, balanced against metallic cations, and a marked increase in oceanic CO_2 therefore requires an increase in cation concentration, which can be brought about only by rock weathering or solution of calcium-rich sediments. These are slow processes.

PROBABLE FUTURE CONTENT OF CARBON DIOXIDE IN THE ATMOSPHERE

We can conclude with fair assurance that at the present time, fossil fuels are the only source of CO₂ being added to the ocean-atmosphere-biosphere system. If this held true throughout the last hundred years, the quantity of CO₂ in the air at the beginning of the present decade was about 7% higher than in the middle of the last century (see Table 3).

Throughout these hundred years, the rate of fossil fuel combustion, and thus of CO₂ production, continually increased, on the average about 3.2 percent per year. The amount produced in 1962 was almost 25 times the annual production in the mid-1860's. The rate of increase may be accelerating. During the eight years from 1954 to 1962, the average rate of increase was 5%.

We can ask several questions about the future CO₂ content of the atmosphere. Two of these questions are:

(1) What will the total quantity of CO₂ injected into the atmosphere (but only partly retained there) be at different future times?

(2) What would be the total amount of CO₂ injected into the air if all recoverable reserves of fossil fuels were consumed? At present rates of expansion in fossil fuel consumption this condition could be approached within the next 150 years.

The second question is relatively easy to answer, provided we consider only the estimated recoverable reserves of fossil fuels. The data are shown in Table 5. We may conclude that the total CO₂ addition from fossil fuel combustion will be a little over 3 times the atmospheric content, and that, if present partitions between reservoirs are maintained, the CO₂ in the atmosphere could increase by nearly 170 percent.

The answer to the first question depends upon the rate of increase of fossil fuel combustion. Table 6 shows that if this combustion remains constant at the 1959 level, the total CO₂ injected into the atmosphere by the year 2000 will be about 28 percent of the atmospheric content in 1950. If the average rate of increase of combustion continues at 3.2 percent per year, the quantity injected into the atmosphere by the year 2000 will be about 42 percent; if the 5% rate of increase during the last 8 years persists, the quantity injected will be close to 60 percent. Assuming further that the proportion remaining in the atmosphere continues to be half the total quantity injected, the increase in amospheric CO₂ in the year 2000 could be somewhere between 14 percent and 30 percent.

Based on projected world energy requirements, the United Nations Department of Economic and Social Affairs (1956) has estimated an amount of fossil fuel combustion by the year 2000 that with our assumed partitions would give about a 25 percent increase in atmospheric CO₂, compared to the amount present during the 19th Century. For convenience, we shall adopt this figure in the following estimate of the effects on atmospheric radiation and temperature.

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Table 5.—Estimated Remaining Recoverable Reserves of Fossil Fuels

	10º Metric Tons	Carbon Dioxide Equivalent, 10 ¹⁸ gms	As % of Atmospheric CO ₂ in 1950
Coal and Lignite ¹	2, 320 212 166 75	5. 88 . 67 . 43 . 24	252 29 18 10
Oil Shales ²	2, 971	7. 85	336

¹ Assumed to be 20 percent lignite containing 45 percent carbon, and 80 percent bituminous coal containing 75 percent carbon.

Source: Computed from data given by M. King Hubbert, "Energy Resources, A Report to the Committee on Natural Resources of the National Academy of Sciences—National Research Council," NAS Publication 1000-D, 1962, pp. 1-141.

Table 6.—Estimates of Carbon Dioxide From Fossil Fuel Combustion in Future Decades, Assuming Different Rates of Increase of Fuel Use

Year	As percent of A	tmospheric CC	₂ in 1950
Growth rate, percent/year	13. 80 17. 30 20. 79 24. 28 27. 77	3. 2 13. 80 18. 00 23. 79 31. 94 41. 96 57. 04	5. 0 13. 80 18. 47 26. 15 37. 90 58. 75 93. 14

POSSIBLE EFFECTS OF INCREASED ATMOSPHERIC CARBON DIOXIDE ON CLIMATE

One of the most recent discussions of these effects is given by Möller (1963). He considers the radiation balance at the earth's surface with an average initial temperature of 15° C (59° F), a relative humidity of 75 percent, and 50% cloudiness. We may compute from his data that with a 25 percent increase in atmospheric CO₂, the average temperature near the earth's surface could increase between 0.6° C and 4° C (1.1° F to 7° F), depending on the behavior of the atmospheric water vapor content. The small increase would correspond to a constant absolute humidity, that is, a constant weight of water in the atmosphere. The larger increase would correspond to a constant relative humidity, that is, as the temperature rose, the water vapor content would also rise to maintain a constant percentage of the saturation value. A doubling of CO₂ in the air, which would happen if a little more than half the reserves of fossil fuels were consumed, would have about three times the effect of a twenty-five percent increase.

As Möller himself emphasized, he was unable to take into account the vertical transfer of latent heat by evaporation at the surface and condensation aloft, or of sensible heat by convection and advection. For this reason he was unable to consider the interactions between different atmospheric layers in a vertical column. In consequence, Möller's computations probably over-estimate the effects on atmospheric temperature of a CO2 increase. A more comprehensive model is being developed by the U.S. Weather Bureau. This includes processes of convection and of latent heat transfer through the evaporation and condensation of water vapor. Meaningful computations should be possible with this model in the very near future. But climatic changes depend on changes in the general circulation in the atmosphere, and these will be related to the spatial distribution and time variation of carbon dioxide and water vapor. The ratio of CO₂ to water vapor is higher in the polar regions than in low latitudes, higher in winter than in summer, and much higher in the stratosphere than near the ground. For example, the volume of carbon dioxide in the atmosphere at high latitudes is about half the volume of water vapor, while near the equator it is less than a tenth of the water vapor volume. As a result, the radiation balance of the earth will be affected differently at different seasons, latitudes, and heights by changes in the atmospheric CO2 content. Without a comprehensive model incorporating both the fluid dynamics and the radiation transfer processes of the atmosphere it is not possible to predict how these effects will perturb the general circulation. Such a model may be available within the next two years.

² Assumed carbon content of petroleum, natural gas liquids, and hydrocarbons recoverable from tar sands and oil shales=86 percent.

 $^{^3}$ Assumed composition of natural gas by volume: CH4=80 percent, $C_2H_6\!=\!15$ percent, $N_2\!=\!5$ percent.

Models of atmospheric thermal equilibrium in which vertical convection is allowed to maintain the observed vertical temperature gradient have recently been constructed by S. Manabe of the U.S. Weather Bureau (Manabe and Strickler, 1964; Manabe, 1965). These show that the effect of infra red absorption from the present atmospheric carbon dioxide at mid latitudes is to maintain a ground temperature about 10°C (18°F) higher than would prevail if no CO₂ were present. An increase in the CO₂ content without a change in absolute humidity would, according to these models, produce a somewhat smaller surface temperature rise than that estimated by Möller. But a considerable change would occur in the stratosphere, where the CO₂ concentration by volume is perhaps 50 times that of water vapor. A 25% rise in carbon dioxide would cause stratospheric temperatures to fall by perhaps 2°C (3.6°F) at an altitude of 30 kilometers (about 100,000 feet) and by 4°C (7°F) at 40 kilometers (about 130,000 feet).

One might suppose that the increase in atmospheric CO2 over the past 100 years should have already brought about significant climatic changes, and indeed some scientists have suggested this is so. The English meteorologist, G. S. Callendar (1938, 1940, 1949), writing in the late 1930's and the 1940's on the basis of the crude data then available, believed that the increase in atmospheric CO2 from 1850 to 1940 was at least 10%. He thought this increase could account quantitatively for the observed warming of northern Europe and northern North America that began in the 1880's. From Table 2 and our estimate of the CO2 partition between the atmospheric, the ocean, and the biosphere, we see that the actual CO2 increase in the atmosphere prior to 1940 was only 4%, at least from fossil fuel combustion. This was probably insufficient to produce the observed temperature changes. [But it should be noted that up to 2.5% of the atmospheric carbon dioxide (after partition with the ocean and the biosphere) could also have been added by the oxidation of soil humus in newly cultivated lands.]

As Mitchell (1961, 1963) has shown, atmospheric warming between 1885 and 1940 was a world-wide phenomenon. Area-weighted averages for surface temperature over the entire earth show a rise in mean annual air temperature of about 0.5°C (0.9°F). World mean winter temperatures rose by 0.9°C (1.6°F). Warming occurred in both hemispheres and at all latitudes, but the largest annual rise (0.9°C or 1.6°F) was observed between 40° and 70° N latitudes. In these latitudes, the average winter temperatures rose by 1.6°C (2.8°F).

The pronounced warming of the surface air did not continue much beyond 1940. Between 1940 and 1960 additional warming occurred in northern Europe and North America, but for the world as a whole and also for the northern hemisphere, there was a slight lowering of about 0.1°C (0.2°F) in mean annual air temperature (Mitchell, 1963). Yet dur-

ing this period more than 40% of the total CO_2 increase from fossil fuel combustion occurred. We must conclude that climatic "noise" from other processes has at least partially masked any effects on climate due to past increases in atmospheric CO_2 content.

OTHER POSSIBLE EFFECTS OF AN INCREASE IN ATMOSPHERIC CARBON DIOXIDE

Melting of the Antarctic ice cap.—It has sometimes been suggested that atmospheric warming due to an increase in the CO₂ content of the atmosphere may result in a catastrophically rapid melting of the Antarctic ice cap, with an accompanying rise in sea level. From our knowledge of events at the end of the Wisconsin period, 10 to 11 thousand years ago, we know that melting of continental ice caps can occur very rapidly on a geologic time scale. But such melting must occur relatively slowly on a human scale.

The Antarctic ice cap covers 14 million square kilometers and is about 3 kilometers thick. It contains roughly 4 x 10¹⁶ tons of ice, hence 4 x 10²⁴ gram calories of heat energy would be required to melt it. At the present time, the poleward heat flow across 70° latitude is 10²² gram calories per year, and this heat is being radiated to space over Antarctica without much measurable effect on the ice cap. Suppose that the poleward heat flux were increased by 10% through an intensification of the meridional atmospheric circulation, and that all of this increase in the flow of energy were utilized to melt the ice. Some 4,000 years would be required.

We can arrive at a smaller melting time by supposing a change in the earth-wide radiation balance, part of which would be used to melt the ice. A 2% change could occur by the year 2000, when the atmospheric CO₂ content will have increased perhaps by 25%. Since the average radiation at the earth's surface is about 2 x 10⁵ gram calories per square centimeter per year, a 2% change would amount to 2 x 10²² calories per year. If half this energy were concentrated in Antarctica and used to melt the ice, the process would take 400 years.

Rise of sea level.—The melting of the Antarctic ice cap would raise sea level by 400 feet. If 1,000 years were required to melt the ice cap, the sea level would rise about 4 feet every 10 years, 40 feet per century. This is a hundred times greater than present worldwide rates of sea level change.

Warming of sea water.—If the average air temperature rises, the temperature of the surface ocean waters in temperate and tropical regions could be expected to rise by an equal amount. (Water temperatures in the polar regions are roughly stabilized by the melting and freezing of ice.) An oceanic warming of 1° to 2°C (about 2°F) oc-

curred in the North Atlantic from 1880 to 1940. It had a pronounced effect on the distribution of some fisheries, notably the cod fishery, which has greatly increased around Greenland and other far northern waters during the last few decades. The amelioration of oceanic climate also resulted in a marked retreat of sea ice around the edges of the Arctic Ocean.

Increased acidity of fresh waters.—Over the range of concentrations found in most soil and ground waters, and in lakes and rivers, the hydrogen ion concentration varies nearly linearly with the concentration of free CO₂. Thus the expected 25% increase in atmospheric CO₂ concentration by the end of this century should result in a 25% increase in the hydrogen ion concentration of natural waters or about a 0.1 drop in pH. This will have no significant effect on most plants.

Increase in photosynthesis.—In areas where water and plant nutrients are abundant, and where there there is sufficient sunlight, carbon dioxide may be the limiting factor in plant growth. The expected 25% increase by the year 2000 should significantly raise the level of photosynthesis in such areas. Although very few data are available, it is commonly believed that in regions of high plant productivity on land, such as the tropical rain forests, phosphates, nitrates and other plant nutrients limit production rather than atmospheric CO₂. This is probably also true of the oceans.

Biological processes are speeded up by a rise in temperature, and in regions where other conditions are favorable higher temperatures due to increased CO₂ might result in higher plant production.

OTHER POSSIBLE SOURCES OF CARBON DIOXIDE

We are fairly certain that fossil fuel combustion has been the only source of CO₂ coming into the atmosphere during the last few years, when accurate measurements of atmospheric carbon dioxide content have been available. Carbon dioxide may have been produced by other sources during earlier times but it is not now possible to make a quantitative estimate. However we can examine the order of magnitude of some of the possible inputs from other sources, on the basis of our knowledge of the processes that might be involved.

Oceanic warming.—The average temperature of the ocean cannot have increased by more than 0.15°C during the past century, since any greater warming would have caused a larger rise in sea level than the observed value of about 10 centimeters. A more probable upper limit is .05°C, because most of the sea level rise can be accounted for by glacial melting. An average .05°C rise would correspond to 0.5° in the top 400 meters. This would cause a nearly 3% rise in the CO₂ partial pressure.

After equilibration with the atmosphere, the partial pressure in both the air and the uppermost ocean layer would be higher by about 2.5%.

Burning of limestone.—Annual world production of carbon dioxide from the use of limestone for cement, fluxing stone, and in other ways, is about 1% of the total from fossil fuel combustion, or 4 x 10⁻⁵ of the atmospheric CO₂ content per year.

Decrease in the carbon content of soils.—Since the middle of the Ninetenth Century, the world's cultivated farmland has been enlarged by about 50%. This is an increase of close to a billion acres or 1.6 million square miles, corresponding to 2.7% of the land area of the earth, and perhaps to 5% of forests and grass lands. Most soil humus is believed to be concentrated in forests and grassy areas. Assume that the total humus is equal to twice the amount of carbon in the atmosphere and that half the carbon in the humus of the newly cultivated lands has been oxidized to carbon dioxide. The total injected into the atmosphere from this source becomes less than 5% of the atmospheric CO_2 .

Change in the amount of organic matter in the ocean.—About 7% of the marine carbon reservoir consists of organic material. Since a 1% change in the carbon dioxide content of the ocean changes the CO₂ pressure by 12.5%, a decrease by 1% in the marine organic carbon (which would increase the total oceanic carbon dioxide by .07%) would raise the carbon dioxide pressure of the ocean and the atmosphere by about 1%. An increase in the temperature of water near the surface, during the past one hundred years, could have speeded up the rate of oxidation of organic matter relative to its rate of production by photosynthesis. Measurements of the content of organic matter in the ocean are neither accurate enough nor sufficiently extended over time to allow a direct estimate of this possibility. A change of several percent could have occurred without detection.

Changes in the carbon dioxide content of deep ocean water.—The deep ocean waters contain about 10% more carbon dioxide than they would if they were at equilibrium with the present atmospheric content. This is a result of the sinking of dead organic remains from the surface waters and their subsequent oxidation in the depths. The combination of biological and gravitational processes can be thought of as a pump that maintains a relatively low carbon dioxide content in the surface waters and in the atmosphere. If the pump ceased to act, the atmospheric carbon dioxide would eventually be increased five fold. Variations in the effectiveness of the pump could have occurred without detection during the past 100 years, and could have caused notable changes in the atmospheric carbon dioxide content.

Changes in the volume of sea water.—During the Ice Age the volume of sea water varied by about 5%. Changes of this magnitude would change the carbon dioxide content of the atmosphere by 10 to 15%.

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But during the last several thousand years, variations in oceanic volume have been small. During the past hundred years, world average sea level has varied by less than 10 centimeters. This very small volume change would have no appreciable effect on the atmospheric carbon dioxide.

Carbon dioxide from volcanoes.—Over geologic time, volcanic gases have been the principal sources of new carbon dioxide injected into the atmosphere. On the average the influx of volcanic CO₂ must have balanced the extraction from the atmosphere by rock weathering. The present rate of influx of volcanic CO₂ is close to a hundred fold less than that from fossil fuel combustion. No data exist on the worldwide level of volcanic activity over geologic time. It is conceivable that the level has fluctuated by two orders of magnitude, and that the fluctuations persisted for millenia, or even for millions of years.

Changes due to solution and precipitation of carbonates.—Calcium and magnesium carbonate precipitation on the sea floor lower the total CO₂ content of ocean water, but increase the carbon dioxide pressure and the free CO₂ content. Conversely, chemical weathering of limestone and dolomite on land lower the atmospheric CO₂ and the free CO₂ content of the sea, but increase the total oceanic CO₂. The rates of these processes are about one order of magnitude lower than the present rate of production of carbon dioxide by fossil fuel combustion.

We conclude that the only sources of carbon dioxide comparable in magnitude to fossil fuel combustion during the last 100 years could have been a decrease in soil humus due to the increase in the area of cultivated lands, a decrease in the content of "dissolved" organic matter in the ocean, or a lowering of the carbon dioxide content of deep ocean waters. Marked changes in the oceanic regime would have been necessary for the latter two processes to have significant effects. As we have shown, none of the three processes are likely to be significant at the present time. Nor are any oceanographic data available which suggest that the required changes in the ocean occurred during the last hundred years.

CONCLUSIONS AND FINDINGS

Through his worldwide industrial civilization, Man is unwittingly conducting a vast geophysical experiment. Within a few generations he is burning the fossil fuels that slowly accumulated in the earth over the past 500 million years. The $\rm CO_2$ produced by this combustion is being injected into the atmosphere; about half of it remains there. The estimated recoverable reserves of fossil fuels are sufficient to produce nearly a 200% increase in the carbon dioxide content of the atmosphere.

By the year 2000 the increase in atmospheric CO₂ will be close to 25%. This may be sufficient to produce measurable and perhaps marked

changes in climate, and will almost certainly cause significant changes in the temperature and other properties of the stratosphere. At present it is impossible to predict these effects quantitatively, but recent advances in mathematical modelling of the atmosphere, using large computers, may allow useful predictions within the next 2 or 3 years.

Such predictions will need to be checked by careful measurements: a series of precise measurements of the CO₂ content in the atmosphere should continue to be made by the U.S. Weather Bureau and its collaborators, at least for the next several decades; studies of the oceanic and biological processes by which CO₂ is removed from and added to the atmosphere should be broadened and intensified; temperatures at different heights in the stratosphere should be monitored on a worldwide basis.

The climatic changes that may be produced by the increased CO₂ content could be deleterious from the point of view of human beings. The possibilities of deliberately bringing about countervailing climatic changes therefore need to be thoroughly explored. A change in the radiation balance in the opposite direction to that which might result from the increase of atmospheric CO2 could be produced by raising the albedo, or reflectivity, of the earth. Such a change in albedo could be brought about, for example by spreading very small reflecting particles over large oceanic areas. The particles should be sufficiently buoyant so that they will remain close to the sea surface and they should have a high reflectivity, so that even a partial covering of the surface would be adequate to produce a marked change in the amount of reflected sunlight. Rough estimates indicate that enough particles partially to cover a square mile could be produced for perhaps one hundred dollars. Thus a 1% change in reflectivity might be brought about for about 500 million dollars a year, particularly if the reflecting particles were spread in low latitudes, where the incoming radiation is concentrated. Considering the extraordinary economic and human importance of climate, costs of this magnitude do not seem excessive. An early development of the needed technology might have other uses, for example in inhibiting the formation of hurricanes in tropical oceanic areas.

According to Manabe and Strickler (1964) the absorption and reradiation of infrared by high cirrus clouds (above five miles) tends to heat the atmosphere near the earth's surface. Under some circumstances, injection of condensation or freezing nuclei will cause cirrus clouds to form at high altitudes. This potential method of bringing about climatic changes needs to be investigated as a possible tool for modifying atmospheric circulation in ways which might counteract the effects of increasing atmospheric carbon dioxide.

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Section II. DETAILED COMPUTATIONS

Calculation of the Relative Sizes of the Ocean and Biosphere Reservoirs and the Probability of Other Sources of Carbon Dioxide at the Present Time

A portion of the carbon dioxide coming into the atmosphere will be transferred to the ocean, and another part will enter the biosphere. We can test the possibility of carbon dioxide sources other than fossil fuel combustion by examining the relative sizes of the required ocean and biosphere reservoirs.

Let

 $A = CO_2$ in atmosphere.

 $B = \text{equivalent CO}_2$ in the part of the biosphere that exchanges with the atmosphere.

 $M=CO_2$ in the oceanic reservoir that exchanges with the atmosphere.

 ΔA , ΔB , ΔM = changes in CO₂ content of these reservoirs.

$$s = \frac{-\Delta C^{14}O_2}{C^{14}O_2}$$
 in the reservoir system, $A + B + M$.

 $a = \frac{\Delta A}{A}$ = fractional change of atmospheric CO₂ content.

 $fA = CO_2$ produced by fossil fuel combustion.

 $bA = CO_2$ produced by other processes.

Then, at equilibrium,

$$\Delta A + \Delta B + \Delta M = (f+b)A$$

$$\Delta A = aA$$

 $\Delta M = oM$, where $o = \frac{a}{12.5}$, owing to the buffer mechanism of sea water (Bolin and Eriksson, 1958).

And assuming that CO_2 produced by other processes has approximately the same C^{14} content as the atmospheric CO_2 , and knowing that fossil fuel CO_2 contains no C^{14} .

$$A+B+M=\frac{f}{s}A$$

Solving for ΔB , M, and ΔM , we find

$$\Delta B = A \left(o + f + b + \sigma c - \frac{of}{s} - a \right) \tag{1}$$

$$M = A\left(\frac{f}{s} - 1 - c\right) \tag{2}$$

$$\Delta M = oA \left(\frac{f}{s} - 1 - c \right) \tag{3}$$

Where

$$c = \frac{B}{A}$$

We know from measurements of tree rings grown during the middle of the Nineteenth Century, compared with those grown during the last few years, that for the period 1850–1950

$$0.02 \ge s \ge 0.01$$
 (the "Suess Effect")

During this same period, f = 0.1, hence

$$\frac{f}{5} \ge s \ge \frac{f}{10}.$$

The series of atmospheric CO₂ measurements at Mauna Loa and Antarctica from 1958–1963 show that during these years

$$a = \frac{f}{2}$$

Substituting in equations (1), (2) and (3) we find, for different values of b and $\frac{B}{A}$, the values shown in Table 7 for $\frac{M}{A}$, $\frac{\Delta M}{f}$ and $\frac{\Delta B}{f}$.

From this table we see that with a "Suess Effect" of 2% (the most probable value), with fossil fuel combustion as the sole source of additional CO₂, and with "effective" biosphere sizes of 2.5 to 0.5 times the atmospheric CO₂, the oceanic reservoir is 2.6% to 6.0% of the volume of the oceans, equivalent to a layer of water 100 to 240 meters thick just below the surface. The size of the oceanic reservoir varies inversely with the size of the biosphere. For a "Suess Effect" of 1% (probably too low), and the same range of biosphere sizes, the assumed layer of complete mixing contains 11% to 14% of the ocean volume, and has an effective thickness of 440 to 560 meters.

Table 7.—Possible Sizes of Oceanic and Biosphere Reservoirs and Partition of Added CO₂ Among Reservoirs

b=0; s=0.	2f; a=0.5f					(f=0.1)	
	$\frac{B}{A}$	$\frac{M}{A}$	$\frac{\Delta B}{f}$	$rac{\Delta M}{f}$	$\frac{\Delta B}{B}$	$rac{\Delta M}{M}$	
1 1 2	0. 5 1. 0 1. 5 2. 0 2. 5	3. 5 3. 0 2. 5 2. 0 1. 5	0. 36 . 38 . 40 . 42 . 44	0. 14 . 12 . 10 . 08 . 06	0. 072 . 038 . 027 . 021 . 018	0. 004 . 004 . 004 . 004 . 004	
b=0; s=0.3							
1 1 2	0. 5 1. 0 2. 5 2. 0 2. 5	8. 5 8. 0 7. 5 7. 0 6. 5	. 16 . 18 . 20 . 22 . 24	. 34 . 32 . 30 . 28 . 26	. 032 . 018 . 013 . 011	. 004 . 004 . 004 . 004 . 004	
b=.5f; s=0.2f; a=0.5f							
1 1 2	0. 5 0 5 2. 0	3. 5 3. 0 2. 5 2. 0 1. 5	. 86 . 88 . 90 . 92 . 94	. 14 . 12 . 10 . 08 . 06	. 172 . 088 . 060 . 046 . 038	. 004 . 004 . 004 . 004	

A layer of water a few hundred meters thick would be acceptable to oceanographers as defining mixing over several decades. Hence, if the "Suess Effect" is close to 2%, the size of the biosphere reservoir is probably about equal to, or less than, that of the atmospheric reservoir. This coincides with other estimates that the effective size of the biosphere on land, including both living organisms and humus, ranges from $\frac{1}{2}$ to one times the atmospheric carbon content. Possibly the organic content of the oceanic mixing layer should be included in the biosphere reservoir, but this is only a few tenths of the atmospheric CO_2 .

In terms of the amounts of carbon they contain, the biosphere reservoir is much smaller than the oceanic reservoir. However, over short times in the ocean, only the relatively small "free" CO₂ content (dissolved and hydrated carbon dioxide) needs to be taken into account. Most of the oceanic carbon dioxide is present as carbonate and bicarbonate, and because of the peculiar buffer mechanism of sea water they do not have a very large quantitative effect on the partition between the sea and the air.

The Table shows if sources other than fossil fuel combustion had contributed much carbon dioxide to the air within the last few decades,

the biosphere would have increased in size by what is probably an observable amount. Such an increase has not been noted. We can conclude that, at least during the recent past, fossil fuel combustion has been the only significant source of CO₂ added to the ocean-atmosphere-biosphere system. The available data do not rule out the possibility that in earlier decades carbon dioxide may have come from oxidation of marine or terrestrial humus, as well as from fossil fuels, but if so, more than half of the amount produced was re-absorbed in the biosphere and the ocean.

REFERENCES

- Arrhenius, Svante, 1903: Lehrbuch der kosmischen Physik 2. Leipzig: Hirzel.
- Bolin, Bert, and Eriksson, Erik, 1958: Changes in the carbon dioxide content of the atmosphere and sea due to fossil fuel combustion. Rossby Memorial Volume, edited by B. Bolin, Rockefeller Institute Press, New York, 1959. pp. 130–142.
- Bolin, B., and Keeling, C. D., 1963: Large-scale atmospheric mixing as deduced from the seasonal and meridional variations of carbon dioxide. *Journal of Geophysical Research*, Vol. 68, No. 13. pp. 3899–3920.
- Broecker, Wallace S., 1963: C¹⁴/C¹² ratios in surface ocean water. Nuclear Geophysics: Proceedings of conference NAS/NRC Publ. 1075, Washington, D.C. pp. 138–149.
- Broecker, Wallace S., 1965: Radioisotopes and oceanic mixing. Manuscript Report, Lamont Geological Observatory, Palisades, New York.
- Brown, Craig W., and Keeling, C. D., 1965: The concentration of atmospheric carbon dioxide in Antarctica. Manuscript submitted for publication, Scripps Institution of Oceanography.
- Callendar, G. S., 1938: The artificial production of carbon dioxide and its influence on temperature. *Quarterly Journal Royal Meteorol. Soc.*, Vol. 64. p. 223.
- Callendar, G. S., 1940: Variations in the amount of carbon dioxide in different air currents. *Quarterly Journal Royal Meteorol. Soc.*, Vol. 66. p. 395.
- Callendar, G. S., 1949: Can carbon dioxide influence climate? Weather Vol. 4. p. 310.
- Callendar, G. S., 1961: Temperature fluctuations and trends over the earth. Quarterly Journal Royal Meteorol. Soc., Vol. 87, No. 371. pp. 1-12.
- Chamberlin, T. C., 1899: An attempt to frame a working hypothesis of the cause of glacial periods on an atmospheric basis. *Journal of Geology*, Vol. 7. pp. 575, 667, 751.
- Conservation Foundation, 1963: Report of conference on rising carbon dioxide content of the atmosphere.
- Craig, Harmon, 1957: The natural distribution of radiocarbon and the exchange time of carbon dioxide between atmosphere and sea. *Tellus*, Vol. 9, No. 1. pp. 1–17.

- Eriksson, Erik, 1963: The role of the sea in the circulation of carbon dioxide in nature. Paper submitted to Conservation Foundation Conference, March 12, 1963.
- Eriksson, Erik, 1963: Possible fluctuations in atmospheric carbon dioxide due to changes in the properties of the sea. *Journal of Geophysical Research*, Vol. 68, No. 13. pp. 3871–3876.
- Hubbert, M. King, 1962: Energy resources; a report to the committee on natural resources of the National Academy of Sciences—National Research Council. Publication 1000-D, National Academy of Sciences— National Research Council, Washington, D.C.
- Kaplan, Lewis D., 1959: The influence of carbon dioxide variations on the atmospheric heat balance. *Tellus*, Vol. 12 (1960), No. 2. pp. 204–208.
- Kraus, E. B., 1963: Physical aspects of deduced and actual climatic change. Annals of New York Academy of Sciences, Vol. 95. pp. 225–234.
- Lamb, H. H., and Johnson, A. I., 1959: Climatic variation and observed changes in the general circulation. *Geografiska Annaler*, Vol. 41. pp. 94–134.
- Lamb, H. H., and Johnson, A. I., 1961: Climatic variation and observed changes in the general circulation. Geografiska Annaler, Vol. 43. pp. 363-400.
- Lieth, Helmut, 1963: The role of vegetation in the carbon dioxide content of the atmosphere. *Journal of Geophysical Research*, Vol. 68, No. 13. pp. 3887–3898.
- Lysgaard, L., 1963: On the climatic variation. *Changes of Climate*, Rome Symposium by UNESCO-WHO; Published in Arid Zone Research Volumes, 20, Paris. pp. 151-159.
- Manabe, Syukuro, and Strickler, Robert F., 1964: Thermal equilibrium of the atmosphere with a convective adjustment. *Journal of Atmospheric Sciences*, Vol. 21, No. 4. pp. 361–385.
- Manabe, Syukuro, 1965: Dependence of the climate of the earth's atmosphere on the change of the content of some atmospheric absorbers. Text of talk made at summer study session of NAS Panel on Weather and Climate Modification.
- Mitchell J. Murray, Jr., 1963: Recent secular changes of global temperature. Annals of New York Academy of Sciences, Vol. 95. pp. 235-250.
- Mitchell, J. Murray, Jr., 1963: On the world-wide pattern of secular temperature change. *Changes of Climate*, Rome Symposium by UNESCO-WHO; Published in Arid Zone Research Volumes, 20, Paris. pp. 161–181.
- Möller, F., 1963: On the influence of changes in the CO₂ concentration in air on the radiation balance of the earth's surface and on the climate. *Journal of Geophysical Research*, Vol. 68, No. 13. pp. 3877–3886.
- Pales, Jack C., and Keeling, C. D., 1965: The concentration of atmospheric carbon dioxide in Hawaii. Manuscript submitted for publication, Scripps Institution of Oceanography.
- Plass, Gilbert N., 1955: The carbon dioxide theory of climatic change. Tellus, Vol. 8 (1956), No. 2. pp. 140–154.

- Plass, Gilbert N., 1956: The influence of the 15μ carbon-dioxide band on the atmospheric infra-red cooling rate. Quarterly Journal Royal Meteorol. Soc., Vol. 82. pp. 310–324.
- Plass, Gilbert N., 1961: Letter to the editor concerning "The influence of carbon dioxide variations on the atmospheric heat balance," by L. D. Kaplan, *Tellus*, Vol. 12 (1960) pp. 204–208. *Tellus*, Vol. 13 (1961), No. 2. pp. 296–300.
- Revelle, Roger, and Suess, Hans E., 1956: Carbon dioxide exchange between atmosphere and ocean and the question of an increase of atmospheric CO₂ during the past decades. *Tellus*, Vol. 9 (1957), No. 1. pp. 18–27.
- Suess, H. E., 1965: Secular variation of the cosmic ray-produced carbon-14 in the atmosphere and their interpretations. Submitted for publication to *Journal of Geophysical Research*, Scripps Institution of Oceanography.
- United Nations Department of Economic and Social Affairs: World energy requirements in 1975 and 2000. Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, 1956. pp. 3–33.
- United Nations, 1961-64: World energy supplies. Statistical Papers, Series J. United Nations, New York.