

CO₂ instruments (left) in the new observatory building 1978, with meteorological and solar radiation strip charts (right).



John Chin with ozonesonde at the National Weather Service airport station, Hilo, in March 1970. The MLO balloon-supported instrument used the standard Weather Bureau radiosonde transmitter and receiver at the airport. 35

Gas measurements

In Mauna Loa Observatory A 20th Anniversary Report John Miller, Editor U. S. Dept. of Commerce September, 1978

THE INFLUENCE OF MAUNA LOA OBSERVATORY ON THE DEVELOPMENT OF ATMOSPHERIC CO₂ RESEARCH

Charles D. Keeling Scripps Institution of Oceanography University of California at San Diego

INTRODUCTION

The increasing amount of CO_2 in the atmosphere from the burning of fossil fuels has become a serious environmental concern. Central to this concern is the question whether a rise in CO_2 constitutes a peril to man by raising world temperatures, as many scientists now claim. That a rise in CO_2 is occurring is unquestionable, however. Mauna Loa Observatory (MLO) data are providing dramatic evidence of that: they show amounts more than 10% over amounts recorded before the Industrial Revolution, and a rise of 6% in the last 19 years alone.

Ninety-seven percent of the energy demand of the industrial world is met today by burning fossil fuels. Even if the industrialized world were to decide to shift to other energy sources as rapidly as possible, the annual consumption of fossil fuels would double before the shift was complete. Without such a shift, a peak annual rate ten or even twenty times today's rate may occur before fuel reserves, especially coal reserves, are exhausted. Thus a large additional increase in atmospheric CO₂ is likely in the next few decades. As Revelle and Suess (1957) wrote, "Through his worldwide industrialized 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 idea that CO_2 from fossil fuel burning might accumulate in air and cause a warming of the lower atmosphere was speculated upon as early as the latter half of the nineteenth century (Arrhenius, 1903). At that time the use of

fossil fuel was too slight to expect a rise in atmospheric CO_2 to be detectable. The idea was again convincingly expressed by Callendar (1938, 1940) but still without solid evidence of a rise in CO_2 .

The first unmistakable evidence of atmospheric CO_2 increase was furnished by continuous measurements made at MLO and by measurements of flask samples collected periodically at the South Pole. These data, obtained in connection with the International Geophysical Year (IGY), were precise enough to indicate a rise in concentration in 1959 when compared with the results of the previous year (Keeling, 1960). Further measurements have shown a persistent year-to-year increase.

Along with new observations have come increasingly refined calculations of the heating effect of increased atmospheric CO_2 . One of the most widely accepted climate models emerging from this effort indicates that the earth's surface would warm by 4°C above the present average global temperature for a fourfold increase in CO_2 , by 6°C for an eightfold increase (Geophysics Study Committee, 1977). A rise in CO_2 as great as eightfold before coal reserves are exhausted has been predicted using a geochemical model calibrated by the Mauna Loa and South Pole trends (Keeling and Bacastow, 1977).

Such a high average global temperature has probably not occurred for tens of millions of years. Accompanying such warming may be shifts in rainfall patterns and in agricultural zones. Polar ice may melt or break up and lead to coastal flooding (Geophysics Study Committee, 1977). These problems, once upon us, will not be easily overcome. Once high CO_2 levels are reached, they will probably decrease only slowly as deep ocean water gradually absorbs the excess CO_2 . Concentrations well above preindustrial levels are likely to persist for at least 1,000 years, along with attending climatic problems (Keeling and Bacastow, 1977).

Whether or not a large CO_2 increase will occur and persist depends on the natural carbon cycle, about which we still know too little. How much CO_2 from fossil fuel will remain in the air during the next centuries? How much will be taken up by the oceans and by vegetation on land? These questions cannot be answered from present knowledge. Sustained monitoring of CO_2 at sites such as MLO is an indispensable aid to validate predictions stemming from calculations of the behavior of the carbon cycle.

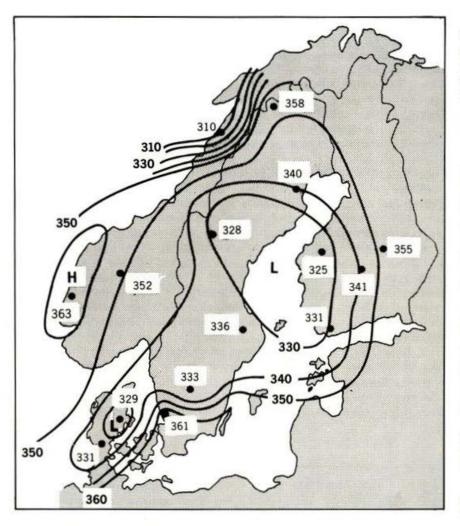


Figure 1. Concentrations of atmospheric CO_2 over Scandinavia (ppm) on February 20, 1955. The approximate pattern is shown by contour lines in a manner similar to Bischof's (1960, Fig. 6). Concentrations were determined by absorption of CO_2 in barium hydroxide solution. Viewed in this context, the reasons for measuring atmospheric CO_2 at Mauna Loa seem compelling. A few of us remember, however, that the original decision to study CO_2 at this remote site was not easily made. Because the story is closely involved with MLO being established in the first place, it seems appropriate to recount here some of the human aspects of this story and its scientific perspective.

HISTORY

The IGY, which began in 1957, offered scientists for the first time an organizational setting for study of atmospheric CO_2 on a global scale. In view of the importance of knowing whether airborne CO_2 was rising worldwide, such a study was long overdue. The data published before the IGY led to a general belief that CO_2 concentrations depended greatly on location with no clear time trend (Bray, 1959). Observations varied from under 200 parts per million (ppm) near the North Pole to over 350 ppm in continental air and air near the equator (Buch, 1948). Owing to this apparent spatial variability, a whole network of stations was deemed necessary to detect any significant global trend.

In the early 1950's, Carl G. Rossby suggested that Stockholm University's Meteorological Institute, which he directed, should participate in an extensive investigation of trace chemicals in the atmosphere as a prelude to the IGY. At a conference held on the subject in 1954, participants decided to plan for a worldwide network of CO₂ monitoring stations, possibly including a site in the Hawaiian islands (Eriksson, 1954). Responsibility for setting up stations in the Pacific region fell to Wendel Mordy, a conference member and chief meteorologist of the Pineapple Research Institute in Honolulu.

When I learned that Mordy was interested in measuring atmospheric CO₂ in the Pacific region, I informed him of CO₂ studies I had begun in 1955 while at the California Institute of Technology. In contrast to previous studies, I had found practically constant atmospheric CO₂ in turbulent air near midday.

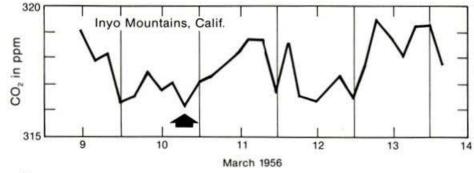
Meanwhile, CO_2 monitoring had just begun in Scandinavia under the general direction of Kurt Buch of Finland. The Scandinavian data (Fig. 1) resembled past work, with greatly varying CO_2 concentrations — even though special care was being taken to sample in open areas away from local influences (Fonselius et al., 1955). My daytime CO₂ results were close to the Scandinavian means, but the variability was far less — even though I had taken special care to sample in densely vegetated areas where local influences would predominate. Specifically, I had found that everywhere I went the air a few tens of meters from the plants on sunny days tended to reach a nearly constant CO₂ level of about 315 ppm (Keeling, 1958). In an attempt to understand why, I took measurements in some exposed windy areas away from plants: at high elevation in the White Mountains (Fig. 2) and Sierra Nevada of California, on ocean beaches, and over ocean water near the equator (Keeling, 1961). All these data were also near 315 ppm. I concluded that the CO₂ in air had a characteristic background concentration, at least near the west coast of the United States and Central America where I had sampled. Evidently, on sunny days this background level prevailed even near plants.

Thus I became concerned that the proposed measurements in Hawaii and elsewhere might not be accurate enough to establish this background CO₂ level. Although Mordy soon decided not to participate in CO₂ studies, my concerns reached the attention of Harry Wexler, Director of Research for the U.S. Weather Bureau. Wexler was a friend of Rossby and an ardent supporter of broadly based meteorological studies. He invited me to Washington early in 1956.

The Weather Bureau already had a small wood frame hut near the summit of Mauna Loa where some simple automatic instruments were housed. In 1955, at Wexler's urging, plans were underway to construct a larger, more permanent structure where people would live and tend more complicated instruments. During my interview with Wexler, which I recall began promptly at 8:00 a.m., I talked to him about the possibility of setting up a continuous recording CO_2 gas analyzer on Mauna Loa since it would be possible to live there and tend the analyzer as necessary. As far as I knew, no one had ever before suggested measuring atmospheric CO_2 continuously. Wexler asked a number of questions in rapid-fire, covering both the scientific and the practical. He was especially interested in costs. We went so far as to discuss setting up a second continuous CO_2 analyzer in Antarctica. Then the interview was over. Altogether it took almost exactly 15 minutes, as scheduled. Wexler had made up his mind to press for CO_2 measurements at Mauna Loa using monies which he hoped would be made available by the participation of the United States in the IGY.

During this same spring of 1956 the oceanographic community was making plans to participate in the IGY. Roger Revelle, as director of the Scripps Institution of Oceanography, was a leader in this effort. Revelle had an intimate knowledge of the natural CO₂ cycle going back to his student days, and he wanted to make sure that man's "vast geophysical experiment" would be properly monitored and its results analyzed. Revelle believed that a CO₂ program should include ocean water studies as well as atmospheric measurements. With this in mind and with Wexler's concurrence, he arranged funding for a laboratory for CO₂ measurements at Scripps, and I was invited to run it. Although it had not been decided precisely what kind of CO₂ program should be implemented as part of the United States IGY effort, I accepted his offer.

Wexler's support of continuous measurements of atmospheric CO_2 at MLO was a bold decision not widely accepted at the time. Wexler knew that I had located a manufacturer of nondispersive infrared CO_2 gas analyzers, but he also knew that I had not yet been able to test such an analyzer. Even the firm itself did not claim that its infrared analyzer was accurate enough for the task.



It had been designed principally for industrial uses which did not demand high accuracy. I was relying on the judgment of one of the firm's engineers that the device was inherently very sensitive and stable. The firm couldn't even lend me one to test. The basic instrument was expensive and required costly additional equipment to operate as an air monitor at a remote field station. Reference gases to calibrate the instrument did not exist.

To most of the IGY planners who heard about the CO_2 infrared analyzer scheme in 1956, such expensive and complicated equipment seemed unnecessary. Both the earlier published data and the new Scandinavian data, appearing in print every 3 months, proved that atmospheric CO_2 variations were so large that traditional methods of chemical analysis would always remain adequate. I distrusted these variable data, but my distrust was based on no more than a few hints from my own data. The most important of these was the near constancy of CO_2 over five days for samples taken at 3,500 m in the White Mountains. Wexler had been especially impressed by the White Mountains record (reproduced in Fig. 2). He felt that if this record was typical of background air, high measurement accuracy at a site on Mauna Loa just might pay off in the IGY program.

Revelle soon agreed to the new infrared analyzer method, but he preferred a network of measuring locations in which such analyzers would be used to analyze air collected in flasks, from ships and aircraft for example.

Rossby remained dubious. I had a chance to meet him just once at an IGY planning meeting at Scripps during 1956. Someone pointed me out to him across a grass lawn during a recess. As he walked up to greet me, he remarked for the benefit of some nearby acquaintances, "Ah . . . za yong man wiz za machine." He seemed upset at this abrupt new American plan to buy expensive gadgetry to measure CO_2 . His skepticism became obvious as we

talked about plans for an ambitious instrument-based United States program.

Ironically, I had so far obtained CO_2 data using quite inexpensive devices — glass sampling flasks, a liquid nitrogen cooled freeze-out trap, a mercury column manometer. But my manometric method could not be used for a large program because a single sample took over an hour to analyze. The infrared gas analyzer was needed to speed up the work without sacrificing high accuracy.

Late in the summer of 1956 I arrived at Scripps to begin implementing the new U.S. atmospheric CO_2 program. In all, four gas analyzers were purchased. One was hastily outfitted for Antarctic field work. Shipment to Little America couldn't be delayed. This first venture turned out, in fact, to be too hasty. No useful data were obtained at Little America until the second Antarctic field year in 1958.

As soon as the Antarctic shipment was off — on the same vessel that was to have carried Admiral Byrd to Antarctica, had he been able to go — I began systematically to test the new analyzers. In March 1957, continuous measurements of air began at Scripps. Soon afterward I assembled another apparatus for Mauna Loa. But there were numerous delays and problems with the aircraft and shipboard programs. These delays were especially bothersome because the IGY had already begun. Soon it would be over, and ships and aircraft would not be available.

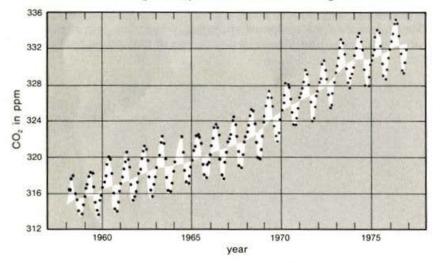
As it turned out, when the equipment for Mauna Loa was ready, I couldn't install it. Revelle insisted that I give first attention to aircraft and shipboard sampling, and the aircraft program was not yet underway. He reinforced his view of the matter by refraining from signing my travel orders to visit Mauna Loa. As the IGY approached its July 1958 ending date, Wexler became very anxious about Mauna Loa. At length he took action himself and

Figure 2. Variation in atmospheric CO_2 over barren ground near White Mountain Research Station in the Inyo Mountains of California during March 1956 (adapted from Fig. 2 of Keeling, 1961). Concentrations were determined manometrically from liquid nitrogen temperature condensates. The arrow identifies the minimum concentration plotted in Fig. 4, accepted as representative of west coast U.S. air.

sent to me Ben Harlin, the meteorologist who had operated the CO_2 equipment at Little America in 1957. With help from Jack Pales, the first director of MLO, Harlin installed the analyzer at MLO in March 1958 without my assistance. To our great surprise, on the first day of operation it delivered within 1 ppm the CO_2 concentration that I had told Harlin to expect on the basis of my earlier manometric data and preliminary test data obtained at Scripps.

Of course this agreement was an accident. The mean of the daytime manometric and Scripps data just happened to be close to the value typical for the month of March. Indeed, the next month's data did not agree — the concentration rose by over one ppm. The following month's mean concentration was still higher. Electrical power failures then shut down the equipment for several weeks. When measuring resumed in July, the concentration had fallen below the March value. I became anxious that the concentration was going to be hopelessly erratic, especially when the computed concentration fell again in late August. Then there were more power shutdowns.

Finally, after my first visit to Mauna Loa in November, the concentration started to climb steadily month by month. Gradually a regular seasonal pattern began to emerge: we were witnessing for the first time nature's borrowing of CO_2 for plant growth during the summer and returning the loan each succeeding winter. Earlier published data for Europe also showed a seasonal trend of sorts (Bray, 1959), but the maximum concentration, arrived at statistically from a highly irregular pattern, was in January, a time of year when CO_2 from burning is likely to accumulate near the ground because of



winter temperature inversions. The maximum at Mauna Loa occurred in May just before temperate and boreal plants add new leaves. The seasonal pattern was highly regular and almost exactly repeated itself during the second year of measurements at Mauna Loa. Thus there was no need to wait for statistical studies to prove the reality of the oscillation as would have been required had less exact chemical methods been used. I soon reviewed my 1955-1956 manometric data and discovered that they showed a similar seasonal variation (Bolin and Keeling, 1963).

No one had expected to determine the long-term rate of rise in CO_2 during the IGY even though establishing the rise was the principal purpose of the program. Revelle and others had expected that the IGY program at best would furnish a reliable "baseline" CO_2 level which could be checked 10 or 20 years later, after the rise in CO_2 was large enough to stand out against local variability. But because of the regularity of the seasonal variation at Mauna Loa a rough estimate of the long-term rise was possible after only two years (Bolin and Keeling, 1963).

Fortunately, funding for CO_2 measurements at MLO was continued after the IGY. By early 1962 it was possible to deduce that approximately half of the CO_2 from fossil fuel was accumulating in the air and that a sink must be carrying a substantial fraction away (Keeling, 1960). Revelle and Suess (1957) had predicted that much of the CO_2 from fossil fuel would be absorbed by the oceans. The earlier published CO_2 data had argued against their view, however, because the rise in CO_2 seemed to be close to that predicted if all of the CO_2 from fossil fuel accumulated in the air. This latter conclusion was reinforced in 1958 after several years of the Scandinavian network data became available (Callendar, 1958). But after four years of measurement at Mauna Loa the question was settled in favor of the Revelle-Suess prediction.

Figure 3. Monthly average concentrations of atmospheric CO₂ at MLO since the beginning of monitoring in 1958. Concentrations were determined with a nondispersive infrared gas analyzer as described by Keeling et al. (1976a), p. 539. As the Mauna Loa record has been further extended, additional interesting features of the long-term trend have revealed themselves. These include perturbations that appear to correlate with the trade winds and with sea surface temperature (Bacastow, 1976; Machta et al., 1976; Newell and Weare, 1977). The seasonal pattern has also been scrutinized to see if variations in amplitude from year to year are meaningful. So far the pattern is too regular to reveal significant variations (Hall et al., 1975). Now after nearly 20 years of measurements, the Mauna Loa record (Fig. 3) appears as a natural yearly cycle gradually being dwarfed by a long-term rise — a dramatic example of inadvertent influence by man on his environment.

THE WEST COAST DATA

Even though the manometric CO_2 data obtained shortly before the IGY played a prominent role in deciding the strategy of the United States CO_2 program, they had never been compared with the infrared CO_2 data for Mauna Loa. Until a pressure broadening correction was recently applied to the latter data (Keeling et al., 1976a), a precise comparison was not possible. It seems worthwhile now to review these earlier measurements and to reconstruct, as closely as possible, the global concentrations of CO_2 back to 1955.

This reconstruction is greatly aided by additional infrared measurements of CO₂ obtained between 1957 and 1962 at La Jolla, California. Although these data were obtained as a by-product of instrument testing, they are nevertheless a useful record of air from the same general geographic area as the earlier manometric data. Except for a few days when air was sampled from a laboratory window, all measurements were made near the end of a 1,000-foot ocean pier where the air was often free of local disturbances, at least during sea breezes. The CO₂ record was twice interrupted for several months when oceanographic work was in progress, but a nearly unbroken continuous record exists from April 1958 to June 1960. Since the Mauna Loa analyzer was operating during this period, these data, and a few more in 1962, are useful in adjusting the La Jolla record to a common basis with Mauna Loa.

Most of the 1955–1956 manometric data reflect local CO₂ emanating from plants and soil. The minimum values for each location, occurring typically near midday, as already noted, may not have been markedly influenced by plant activity, however. A plausible reason for this is that the sampling locations I had chosen were in wild areas which had never been disturbed very much by humans. In wild areas the photosynthetic withdrawal of atmospheric CO₂ by the plants and the release of CO₂ by plant respiration and decomposition should not differ greatly. The net change in the CO₂ concentration of the local air should therefore be relatively small, especially if air turbulence, typically maximal at midday, further diminishes the net effect.

At several control sites on ocean beaches and barren mountains, where I also sampled during 1955 and 1956, the CO₂ concentrations usually agreed with the minimum values found near plants. For example, in Yosemite National Park in June 1955, the lowest value found for forest air was 316.2 ppm; a few miles away over barren terrain near Lake Tenaya, I found 315.9 ppm (Eriksson, 1954).

The minimum CO_2 concentrations for all CO_2 sites in the western United States are listed in Table 1 and plotted in Fig. 4, except that data have been omitted if the humidity was not measured, since for these data it is impossible to determine the CO_2 concentration versus dry air. Most of the measurements were obtained in California, but a few were obtained farther north in the state of Washington and several from Arizona.

Table 1. Minimum atmospheric carbon dioxide concentrations (relative
to dry air) by direct manometric analysis, for various sites near the
west coast of the United States and Central America

Location	Elevation (above sea level)	Date	Local Time	Minimum CO ₂ Concentration (ppm)	CONTRACTOR OF A DECEMPTOR OF A
1955		- 9 - 14 - 14 - 14 - 14 - 14 - 14 - 14 -	All mark		
Big Sur State Park (36 °N., 122 °W.)	70 m	May 18	12:15	319.3	forest
Yosemite National Park	2500 m	June 2	12:30	316.2	forest
(38°N., 119°W.)	3000 m	June 3	10:00	315.9	barren ground
Olympic National Park	170 m	Sept. 7	13:30	312.6	forest
(48°N., 124°W.)	0 m	Sept. 7	15:15	313.8	ocean beach
Gulf of Tehuantepec				314.4	over ocean
(9°N., 89°W.)	10 m	Dec. 1	5:30	314.9*	
1956					
Borrego Valley, California (33 °N., 116 °W.)	340 m	Feb. 1	10:30	314.1	barren desert
Inyo Mountains, California (37°N., 118°W.)	3800 m	Mar. 10	20:00	316.2	barren snow field
Organ Pipe Cactus National Monument (32 °N., 113 °W.)	550 m	Apr. 22	0:00	316.1	desert brush
Howard Pocket, Arizona (35°N., 112°W.)	2100 m	May 16	15:30	317.4	forest
Telephone Hill, Arizona (37°N., 112°W.)	2600 m	May 18	15:00	320.0	forest
Big Sur State Park (36 °N., 122 °W.)	70 m	June 6	12:00	318.4	forest
Yosemite National Park (38 °N., 119 °W.)	2500 m	June 11	12:00	316.4	forest

*Adjusted to 33 °N.

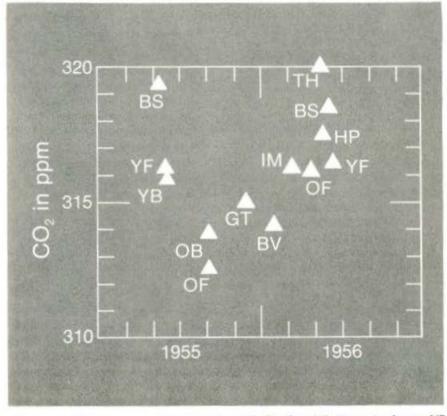


Figure 4. Minimum concentrations of atmospheric CO₂ at various sites near the west coast of the United States during 1955 and 1956. Concentrations were determined manometrically from liquid nitrogen temperature condensates. Sites are identified as follows: BS, Big Sur; YF, Yosemite forest; YB, Yosemite barren ground; OF, Olympic forest; OB, Olympic beach; GT, Gulf of Tehuantepec; BV, Borrego Valley; IM, Inyo Mountains; OP, Organ Pipe; HP, Howard Pocket; TH, Telephone Hill. Also, as a single exception to the above site distribution, Table 1 includes the minimum CO₂ concentration from a suite of samples collected aboard ship off the coast of Nicaragua near 9°N, in 1955. This minimum has been adjusted upward by 0.5 ppm on the basis of the average latitudinal gradient found by Bolin and Keeling (1963) between 9° and 33°N for the appropriate month of sampling.

The continuous measurements obtained at La Jolla from 1957 to 1962 are highly contaminated by local and regional urban sources of CO₂. Even the daily minima, which usually occurred during sea breezes, vary considerably depending on the history of the air. Highest values typically occurred when the air had previously passed near the city of Los Angeles to the northwest. To reduce further the influence of contamination, the daily minima were arranged into calendar weeks, and weekly minima were identified. As noted already in 1960 (Keeling, 1961), these weekly minima scatter much less than the dailies. Also, unlike the dailies their monthly means show a consistent trend suggestive of uncontaminated air.

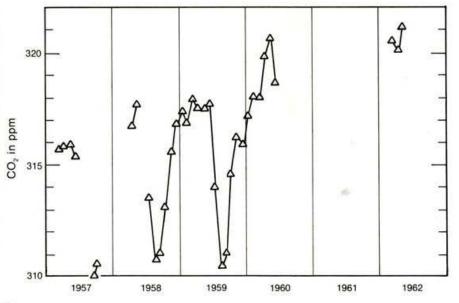
These monthly means are listed in Table 2 and plotted in Fig. 5. One entry, for June 1958, is omitted from further consideration because only one weekly minimum was obtained that month. Also, as indicated in the table, a few obviously contaminated minima were omitted in assembling the monthly means. The means for April 1958 through March 1960 have been published (Keeling, 1961). These, and previously unpublished data for 1957, 1960, and 1962, are here reported according to the 1974 manometric CO₂ mole fraction scale, using formulas for conversion from an adjusted index scale (Keeling et al., 1976a).

The manometric and infrared data (Figs. 4 and 5) display a seasonal variation similar to but of greater amplitude than that for Mauna Loa. The

Average date	No. of weekly minima included	Average observed CO ₂ concentration (ppm)
1957-Mar. 24		- industrial and a state
Apr. 17	4	315.91
May 16	4	315.92
June 16	4	315.43
Sept. 12	5	310.16
Oct. 4	2	311.15
1958-Apr. 21	3*	316.72
May 16	3	317.71
June 22	1	319.08*
July 17	5	313.52
Aug. 18	4	310.83
Sept. 13	4	311.08
Oct. 19	3*	313.17
Nov. 20	5	315.64
Dec. 20	4	316.76
1959–Jan. 19	3*	317.38
Feb. 14	4	316.89
Mar. 18	5	317.89
Apr. 18	4	317.52
May 17	4	317.52
June 14	4	317.65
July 14	5	313.95
Aug. 16	4	310.52
Sept. 12	4	311.14
Oct. 13	5	314.58
Nov. 8	2*	316.19
Dec. 16	5	315.95
1960-Jan. 16	3*	317.19
Feb. 14	4	317.94
Mar. 17	5	317.95
Apr. 16	4	319.82
May 12	3	320.57
June 12	2	318.58
1962-Mar. 25	2	320.43
Apr. 13	4	320.06
May 6	2	321.07

Table 2. Mean of weekly minimum concentration of atmospheric carbon dioxide (relative to dry air) by infrared gas analysis, for Scripps pier, La Jolla, California, at 33°N, 117°W, elevation 8 m

> Figure 5. Monthly averages of the weekly minimum atmospheric CO₂ concentration at La Jolla, California. Concentrations were determined with a nondispersive infrared gas analyzer.



*One weekly minimum omitted from average.

Table 3. Adjusted manometric data,	1955-1956,	and	infrared	gas	analyzer
data, 1957					

		Concentra	Concentration (ppm)			
Location	Month and Year	Adjusted to 15th of the month	Adjusted for seasonal variation	Departure from linear trend		
Big Sur State Park Yosemite National Parl	May 1955	319.36*	316.40	3.46		
at 3000 m	June	315.05	313.72	.72		
Olympic National Park						
at 170 m	Sept.	312.81*	317.16	3.98		
Gulf of Tehuantepec	Dec.	315.36	313.94	0.59		
Borrego Valley	Feb. 1956	314.06	312.92	-0.55		
Inyo Mountains	Mar.	316.35	314.75	1.23		
Organ Pipe Cactus						
National Monument	Apr.	315.91	313.23	-0.35		
Howard Pocket	May	317.42	314.46	0.82		
Telephone Hill	May	320.06*	317.10	3.46		
Big Sur State Park	June	317.74*	316.41	2.71		
Yosemite National Parl	ĸ					
at 2500 m	June	316.08	314.75	1.05		
La Jolla	Mar. 1957	315.46	313.86	-0.36		
La Jolla	Apr.	315.85	313.17	-1.11		
La Jolla	May	315.94	312.98	-1.35		
La Jolla	June	315.51	314.18	-0.21		
La Jolla	Sept.	310.26	314.61	0.16		
La Jolla	Oct.	312.03	314.47	-0.04		

*Judged to be contaminated.

seasonal variation, however, is clearly evident only for the La Jolla data because the 1955–1956 manometric data involve so many missing months and extend over less than two years.

Several of the manometric data appear to be inconsistent with the seasonal trend. That the two CO_2 minima for Big Sur State Park may be too high, both in 1955 and 1956, is not surprising because sampling was done in a public campground where daytime automobile traffic may have produced several ppm of contamination. Also, the CO_2 minimum for Telephone Hill, Arizona, seems too high relative to Howard Pocket, but there is no obvious reason, since the site was in a remote forest north of the Grand Canyon. Finally, the pair of CO_2 minima for the Olympic National Park agree with each other but are both considerably higher than had been expected for the month of sampling on the basis of the La Jolla data, again for no obvious reason.

Before deciding on the disposition of these possibly contaminated values, an adjustment of the data was made to the 15th of the month of sampling in order to reduce scatter resulting from uneven spacing in time. The adjustments were made following a procedure described previously (Keeling et al., 1976b). Specifically, the individual monthly concentrations X(t), in ppm, where t denotes the time in years after January 1, 1955, were fit by the method of least squares to an oscillatory-linear trend function:

$$X(t) = Q_1 \sin 2\pi t + Q_2 \cos 2\pi t + Q_3 \sin 4\pi t +$$
(1)

$$Q_4 \cos 4\pi t + Q_5 + Q_6 t$$

The four possibly contaminated data mentioned above were tentatively omitted from the computation. The parameters of best fit were found to have the values:

$Q_1 =$	2.86883 ppm	$Q_4 = 6.64806 \text{ ppm}$	
$Q_2 =$	0.879716 ppm	$Q_5 = 312.684 \text{ ppm}$	(2)
$Q_3 =$	-1.51123 ppm	$Q_6 = 0.6954 \text{ ppm yr}^{-1}$	

On the basis of equations (1) and (2), the data, including the tentatively rejected values, were adjusted to the 15th of the month as listed in Tables 3 and 4. Next, the data were seasonally adjusted using the first four terms of

613

Month	Concentration at La Jolla*	Trend at Mauna Loa**	Difference		
Apr. 1958	316.54	315.34	1.20		
May	317.73	315.40	2.33		
June		315.47			
July	313.72	315.53	-1.81		
Aug.	310.96	315.59	-4.63		
Sept.	311.15	315.64	-4.49		
Oct.	312.82	315.69	-2.87		
Nov.	315.30	315.74	-0.44		
Dec.	316.67	315.78	0.89		
Jan. 1959	.317.42	315.83	1.59		
Feb.	316.89	315.88	1.01		
Mar.	317.79	315.93	1.86		
Apr.	317.43	315.99	1.44		
May	317.56	316.05	1.51		
June	317.57	316.13	1.44		
July	313.85	316.20	-2.35		
Aug.	310.57	316.29	-5.72		
Sept.	311.24	316.38	-5.14		
Oct.	314.75	316.46	-1.71		
Nov.	316.72	316.55	0.17		
Dec.	315.93	316.63	-0.70		
Jan. 1960	317.20	316.70	0.50		
Feb.	317.94	316.77	1.17		
Mar.	317.88	316.84	1.04		
Apr.	319.79	316.90	2.89		
May	320.52	316.96	3.56		
June	318.34	317.01	1.33		
Mar. 1962	320.08	318.37	1.71		
Apr.	320.13	318.43	1.70		
May	321.02	318.49	2.53		

Table 4. Comparison of atmospheric carbon dioxide concentrations at La Jolla with the long-term trend in concentration at MLO

*Adjusted to the 15th of the month.

**Determined for the 15th of the month from a spline fit of the seasonally adjusted monthly means for 1958–1976, inclusive. equation (1), and the resulting trend data were plotted as shown in Fig. 6. From this plot it becomes clear that the questionable values, shown as crosses, should be rejected. A statistical computation bears this out: the four values differ by factors of 3.4 to 4.9 times the root mean square departure of the remaining 13 data points for 1955–1957 with respect to equations (1) and (2).

The next step was to establish from overlapping data the difference in seasonal variation and long-term trend for Mauna Loa and La Jolla. First, from the entire Mauna Loa record of monthly averages from March 1958 through December 1976, the average seasonal variation and seasonally adjusted trend for that station were established.

Several methods have been used previously to separate the long-term trend at Mauna Loa from the associated seasonal variation (Bacastow, 1977). Here I have chosen to express the trend by a cubic spline function (Reinsch, 1967) and the seasonal variation as an average of the monthly mean concentrations after subtracting the trend. Since the two features are not uniquely separable, an iterative procedure was used. First, an estimate of the long-term trend was found assuming a linear increase with time, and a preliminary estimate of the seasonal variation was obtained. Then consistent with this seasonal variation, the original monthly values were seasonally adjusted, and a cubic spline function was passed through the adjusted data points. Further iterations were carried out until the adjusted values approached constancy. This convergence was rapid, and because of the high regularity of the seasonal variation, the seasonal variation found was similar to that found by using a least squares fit based on equation (1).

Next, as shown in Table 4, the long-term trend for Mauna Loa, expressed as a spline function, was compared with the La Jolla data adjusted to the 15th of each month. For the relatively short period of the comparison it seems reasonable to assume that the long-term trends for Mauna Loa and La Jolla differ by only a constant. On the basis of the monthly differences between the Mauna Loa trend and the La Jolla data (last column of Table 4), mean differences between stations were determined for each month. The sum of these differences is -0.42 ppm; that is, the La Jolla weekly minima, on average, are lower by that amount than the Mauna Loa trend. Since the expected latitudinal difference between stations according to aircraft and shipboard data analyzed by Bolin and Keeling (1963) is -0.20 ppm, the weekly minima agree closely with expectations in spite of the high degree of selection involved in obtaining them. Evidently, the large irregular variations in the original La Jolla record are almost solely owing to high values, probably produced by urban sources.

Next, from the west coast data, 1955-1962, a long-term trend and an average seasonal variation were found in the same manner as that just

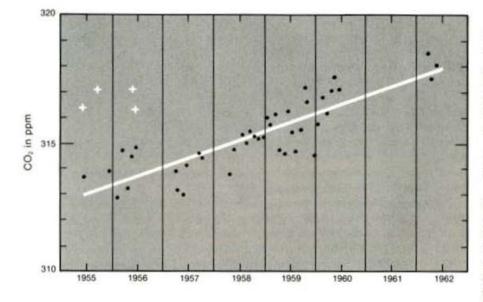


Figure 6. Long-term trend in the minimum concentration of atmospheric CO₂ near the west coast of the United States based on the data of Figs. 4 and 5 after adjustment of each point to the 15th of the month of observation. The seasonal variation. expressed by the first four terms of equation (1) as a harmonic function with 6- and 12-month terms, was subtracted to obtain seasonally adjusted concentrations shown as dots. Possibly contaminated data are indicated by crosses. The straight line is a least-squares best fit through the plotted points.

described for the Mauna Loa record. Because of the considerable gaps in the data, the trend in all iterations was assumed to be a straight line. The final trend, shown in Fig. 7, obeys the relation

$$X(t)_{\text{seasonally adjusted}} = Q_5 + Q_6 t \tag{3}$$

where

$$Q_5 = 312.592, Q_6 = 0.7167 \text{ ppm yr}^{-1}$$
 (4)

and, again, t = 0 for January 1, 1955.

The corresponding seasonal variation, shown in the third column of Table 5, agrees closely with that obtained (see the second column) by comparing the La Jolla data for 1958–1962 with the Mauna Loa spline function trend. The only month where the agreement is possibly unsatisfactory is December which includes the data point from 9°N. This discrepancy does not appear to be significant, however, in view of the scatter of the other 1955–1956 data.

Evidently it makes little difference which seasonal variation is used in further analysis. Since the seasonal variation based on the entire data set from 1955 to 1962 results in slightly lower scatter, I chose this representation.

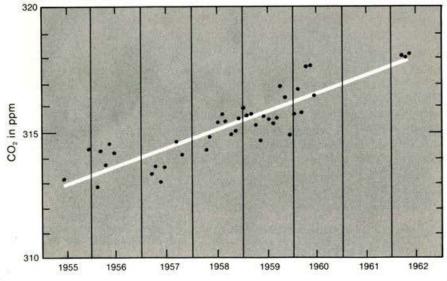
To express the comparison of the pre-1958 U.S. data with the Mauna Loa record, I have devised what I call "proxy" data. My goal is to produce, with the least interpretive adjustment, a set of monthly values valid for Mauna Loa for 1955 through 1957. On the basis of the difference between the seasonal variations for Mauna Loa and the west coast U.S. data for 1955-1962, with due regard for the average difference of 0.42 ppm between locations (Table 6 and Fig. 8), the west coast U.S. data were converted to equivalent Mauna Loa monthly means. In this way the scatter is included, and no judgment of the long-term trend is placed on these early measurements. The results are listed in Table 7 and plotted in Fig. 9. Finally, a long-term trend line was established for the seasonally adjusted Mauna Loa record including these proxy data (Table 8 and Fig. 10). The previous iterative method was again used to separate the trend from the seasonal variation. Since the Mauna Loa record already includes 19 years of direct data, the new data have a negligible effect on the computed seasonal variation for Mauna Loa, shown in the third column of Table 6. Also, since the spline function at any part of the record is

Month	La Jolla 1958-1962*	West Coast 1955-1962**	Difference
Jan.	1.47	1.46	.01
Feb.	1.51	1.22	.29
Mar.	1.96	2.05	09
Apr.	2.23	2.18	.05
May	2.90	2.87	.03
June	1.81	1.87	06
July	-1.66	-1.70	.04
Aug.	-4.76	-4.79	.03
Sept.	-4.40	-4.37	03
Oct.	-1.87	-2.11	.24
Nov.	0.28	0.28	.00
Dec.	0.52	1.04	52
			<i>σ</i> = .20

Table 5. Seasonal variation in atmospheric carbon dioxide near U.S. west coast determined by summing monthly concentrations with the long-term trend removed

*Monthly means of the fourth column entries of Table 4 normalized by adding 0.42 ppm to each value.

**Based on comparison with the linear trend for the west coast of the United States, expressed by equation (3).



sensitive only to nearby data, the inclusion of the early data affects the trend line only near its former beginning in 1958.

The small difference of 0.42 ppm between the La Jolla and Mauna Loa trends where they overlap suggests that the La Jolla weekly minima are not biased, but actually one need not make this assumption in accepting the proxy data, provided that the original west coast minima for 1955–1957 have the same bias as those for 1958–1962. This appears reasonable for 1957 because the data are for the same location as the 1958–1962 data and were selected in the same way. Indeed, as can be seen from Fig. 10, the seasonally adjusted proxy data for 1957 appear to be consistent with the direct data (1958 and later) both as to scatter and trend. Thus one is encouraged to accept the 1957 proxy data as reliable.

One is less confident that the 1955–1956 proxy data are unbiased. Their scatter is greater, and a backward extrapolation of the relatively steep trend line for 1958 suggests that they could be too high by as much as 1.0 ppm. On the other hand, the rise and fall in trend indicated by the spline function for 1955–1956 (Fig. 10) is similar to abrupt changes in trend that have occurred more recently, for example in 1973. Thus one cannot easily decide that the proxy data for 1955–1956 are wrong.

We are probably expecting too much to consider that these early data might tell us something about a change in the long-term trend. These data are better regarded as the kind of "baseline" data which Revelle had in mind to obtain during the IGY. At least they add evidence that no very unusual circumstances influenced the atmospheric CO_2 record immediately before systematic data collecting began during the IGY.

Figure 7. Long-term trend in atmospheric CO_2 for the west coast of the United States, as in Fig. 6 except that the seasonal variation was determined as an average of the monthly average concentrations after subtracting a linear estimate of the trend. Data identified in Fig. 6 as possibly contaminated are not shown.

Month	West Coast*	Mauna Loa	Difference
lan.	1.02	-0.15	-1.17
Feb.	0.80	0.53	-0.27
Mar.	1.63	1.21	-0.42
Apr.	1.76	2.27	0.51
May	2.45	2.74	0.29
June	1.45	2.24	0.79
July	-2.12	0.87	2.99
Aug.	-5.12	-1.12	4.09
Sept.	-4.79	-2.68	2.11
Oct.	-2.53	-2.99	-0.46
Nov.	-0.14	-1.96	-1.82
Dec.	0.62	-0.97	-1.59

٠

Table 6. Seasonal variation in atmospheric carbon dioxide — comparison of west coast United States, 1955-1962, with MLO, 1958-1975

*Third column of Table 5 reduced by 0.42 ppm.

Figure 8. Atmospheric CO_2 as a function of the month of the year determined as a departure of the monthly mean concentration from the long-term trend for Mauna Loa. Data are shown for MLO by dots, and for the west coast of the United States by crosses. Months 1 to 6 (January through June) are plotted twice to reveal the seasonal patterns more fully.

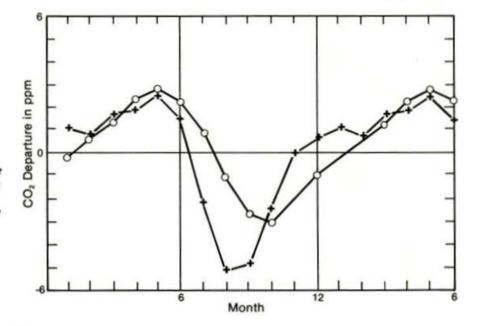


Figure 9. Trend in atmospheric CO_2 concentrations at MLO. The dots indicate the monthly average concentration. Data in 1955, 1956, and 1957 are proxy data based on observations for the west coast of the United States. The oscillatory curve is a spline fit of the sum of the long-term trend and the average seasonal variation determined as in Fig. 7.

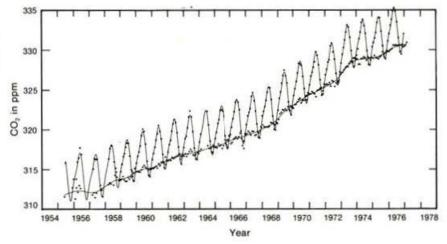


Table 7. Monthly average concentration of atmospheric carbon dioxide (ppm) at MLO expressed according to the 1974 manometric mole fraction scale

See 14	Jan.	Feb.	Mar.	Apr.	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.
1955						315.84*						313.77*
1956		313.79*	315.93*	316.42*	317.71*	316.87*						
1957			315.04*	316.36*	316.23*	316.30*			312.37*	311.57*		
1958			316.33	317.59	317.93	317.71	315.92	315.15	314.02	312.83	313.64	314.71
1959	315.62	316.59	316.94	317.77	318.29	318.24	316.67	314.96	314.12	313.58	315.14	315.77
1960	316.62	317.16	317.90	319.21	320.02	319.74	318.15	316.00	314.23	314.07	315.04	316.19
1961	316.97	317.74	318.63	319.43	320.47	319.71	318.78	316.84	315.16	315.56	316.14	317.13
1962	318.06	318.59	319.74	320.63	321.21	320.83	319.55	317.75	316.27	315.62	316.84	317.70
1963	318.80	319.08	320.15	321.49	322.25	321.50	319.67	317.61	316.25	316.17	317.01	318.36
1964	319.37					322.19	320.49	318.48	317.13	317.02	317.84	318.78
1965	319.55	320.65	321.15	322.31	322.35	322.19	321.53	319.13	317.99	317.70	319.15	319.27
1966	320.22	321.23	322.13	323.30	323.57	323.29	322.36	319.71	317.89	317.54	319.36	320.51
1967	321.60	322.03	322.50	324.00	324.46	323.46	322.19	320.57	318.91	318.81	320.24	321.59
1968	322.15	322.73	323.50	324.52	325.11	325.06	323.62	321.55	319.89	319.80	320.73	322.25
1969	323.73	324.53	325.62	326.58	327.24	326.53	325.63	323.28	322.21	321.67	322.61	324.07
1970	324.91	325.81	326.85	328.07	327.97	327.77	326.44	324.92	323.49	323.50	324.34	325.39
1971	326.46	326.93	327.56	328.23	329.51	329.04	327.87	326.00	324.06	324.20	325.48	326.62
1972	327.30	328.20	328.50	330.22	330.58	329.48	328.56	326.77	325.39	325.72	326.97	328.09
1973	329.16	330.02	330.95	331.95	332.85	332.58	331.30	329.64	328.12	327.67	328.69	329.05
1974	329.84	331.13	331.93	333.16	333.53	332.73	331.77	329.63	327.87	327.84	328.77	330.12
1975	330.64	331.20	331.89	333.14	333.78	333.75	332.06	330.25	328.85	328.58	329.61	330.82
1976	331.75	332.81	333.55	334.62	335.01	334.58	333.22	331.24	329.48	329.19	330.35	331.72

*Proxy data.

Figure 10. Long-term trend in atmospheric CO_2 concentration at MLO. The plot is the same as Fig. 9 except that the seasonal variation has been subtracted out.

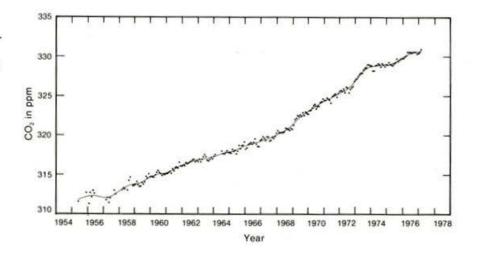


Table 8. Seasonally adjusted concentration of atmospheric carbon dioxide (ppm) at MLO for the 15th of each month expressed according to the 1974 manometric mole fraction scale*

1	Jan.	Feb.	Mar.	Apr.	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.
1955						313.81	313.88	313.96	314.03	314.10	314.16	314.21
1956	314.25	314.28	314.31	314.33	314.34	314.33	314.31	314.28	314.24	314.21	314.17	314.13
1957	314.11	314.09	314.09	314.12	314.16	314.22	314.30	314.39	314.50	314.60	314.71	314.83
1958	314.94	315.05	315.15	315.26	315.36	315.46	315.55	315.63	315.69	315.74	315.77	315.79
1959	315.82	315.85	315.88	315.93	315.99	316.07	316.16	316.26	316.37	316.48	316.58	316.67
1960	316.76	316.83	316.90	316.96	317.01	317.05	317.08	317.11	317.13	317.15	317.18	317.22
1961	317.27	317.33	317.40	317.48	317.56	317.66	317.75	317.84	317.94	318.02	318.10	318.18
1962	318.25	318.32	318.39	318.45	318.51	318.57	318.63	318.68	318.73	318.76	318.80	318.84
1963	318.87	318.91	318.94	318.98	319.01	319.03	319.05	319.08	319.12	319.16	319.21	319.27
1964	319.33	319.39	319.44	319.50	319.56	319.61	319.66	319.70	319.74	319.78	319.82	319.86
1965	319.90	319.95	320.00	320.06	320.12	320.20	320.28	320.36	320.43	320.50	320.57	320.63
1966	320.69	320.75	320.80	320.85	320.90	320.94	320.98	321.03	321.08	321.13	321.19	321.25
1967	321.32	321.38	321.43	321.48	321.54	321.59	321.65	321.73	321.80	321.89	321.97	322.06
1968	322.14	322.22	322.30	322.39	322.47	322.57	322.68	322.80	322.93	323.08	323.24	323.42
1969	323.60	323.78	323.94	324.10	324.24	324.38	324.50	324.62	324.73	324.83	324.94	325.05
1970	325.16	325.28	325.38	325.49	325.60	325.71	325.82	325.93	326.03	326.13	326.22	326.30
1971	326.38	326.45	326.52	326.60	326.68	326.77	326.86	326.95	327.04	327.13	327.22	327.30
1972	327.39	327.47	327.56	327.65	327.76	327.89	328.03	328.20	328.39	328.59	328.81	329.02
1973	329.23	329.45	329.63	329.82	329.99	330.14	330.27	330.37	330.45	330.50	330.54	330.57
1974	330.59	330.62	330.64	330.66	330.68	330.69	330.70	330.72	330.73	330.75	330.78	330.81
1975	330.85	330.90	330.96	331.03	331.11	331.20	331.29	331.40	331.50	331.61	331.72	331.82
1976	331.93	332.02	332.10	332.17	332.23	332.27	332.31	332.34	332.37	332.40	332.44	332.47

*Entries before March 1958 are based on proxy data.

51

EPILOGUE

Since these proxy data for Mauna Loa were originally obtained from sampling sites presumed to be disturbed locally, it seems paradoxical that truly reliable data were not obtained by investigators who deliberately sought undisturbed locations to obtain baseline CO₂ data. As Bray (1959) noted, several nineteenth-century investigators, who claimed analytical analyses accurate to 1.0 ppm, made serious attempts to obtain data representative of locally undisturbed air. I conclude that these scientists, perhaps from an inadequate knowledge of meteorology and atmospheric motion, underestimated the difficulty in finding truly uncontaminated sites. When their analytical and sampling methods failed to give them the high reproducibility that they thought they had attained, they ascribed the scatter to the atmosphere itself and not to weaknesses in their methods.

In the first half of this century declining interest in atmospheric CO_2 was kept alive by only a few investigators. The most notable was Kurt Buch of Finland, who concluded after many years of study that the CO_2 concentration varied systematically with air mass. His claims (Keeling and Bacastow, 1977) that high arctic air had concentrations in the range of 150 to 230 ppm, north and middle Atlantic air, 310 to 345 ppm, and tropical air, 320 to 370 ppm, strongly influenced preparations for the IGY CO_2 program, especially the Scandinavian program, which he initially supervised. When from inadequate chemical and sampling techniques the Scandinavian pre-IGY program produced CO_2 concentrations in the same range as previous data, these new data were readily justified as resulting from different properties of the air masses passing over the sampling sites (Fonselius et al., 1956).

How long would the findings of the Scandinavian CO₂ network have been accepted if new manometric and infrared studies had not been begun? The Scandinavian data continued to appear in the back pages of *Tellus* until after the infrared analyzer results for Mauna Loa and other locations had been presented at the International Union of Geodesy and Geophysics meeting in Helsinki in 1960. But reform was on the way. Walter Bischof in 1959 had assumed responsibility for Swedish measurements. He soon became suspicious of their variability on the basis of discrepancies between ground-level and aircraft sampling (Bischof, 1960). Also, he had begun to use an infrared gas analyzer. With this abandonment of the traditional chemical method of analysis, the Swedish CO₂ data ceased to include unreasonably low CO₂ values. Then in 1960 Bischof turned to investigating suspiciously high values using aircraft to verify ground-level data. Probably within a year or two, considerably more accurate systematic data would have begun to appear from the Scandinavian program.

But it is far from certain that a Scandinavian site as reliable as MLO would have soon been established. The Scandinavian investigators lacked the funds to embark on an ambitious continuous sampling program at a remote station. Many years might have passed before data of the guality of the Mauna Loa record would have been forthcoming. Indeed, high costs almost caused MLO to close down in 1964 in spite of its obvious value as a CO₂ sampling site. Disruptions under that threat of closure account for a serious gap in the CO₂ record during the early part of 1964. Problems of cost also contributed to the decision to shut down the South Pole continuous-analyzer program at the end of 1963. If these two remarkable sites had not already been established and yielded high-quality data before 1964, it is likely that the stimulus to start work at such remote sites would not have occurred for at least several more years because of financial impediments. Thus it was a fortunate circumstance that Wexler and Revelle in 1956 saw the value of using the IGY organization to check out the possibility of near constancy in atmospheric CO2 by inaugurating a precise sampling program. We all recognize now that such a program is essential if we are to document adequately the rise in atmospheric CO2.

ACKNOWLEDGMENTS

Many people who could not be included in the historical discussion contributed to the planning of atmospheric carbon dioxide measurements at Mauna Loa. I am particularly indebted to Oliver Wulf, U.S. Weather Bureau scientist stationed at the California Institute of Technology in 1955 and 1956. Wulf first brought my manometric work to the attention of Dr. Wexler. I am also indebted to Paul Humphrey, Dr. Wexler's assistant, who coached me on making the best use of the short time I would have to talk with Wexler. Humphrey later devoted many hours to coordinating funding and logistics involved in setting up CO₂ research at Mauna Loa.

In addition, I am indebted to Kenyon George, engineer of the Applied Physics Corporation, Pasadena, California. George patiently replied to my detailed questions during 1956 about the performance characteristics of his firm's nondispersive infrared gas analyzer. He was not himself convinced that atmospheric CO_2 could be determined by infrared analysis to the accuracy I sought, but his frank answers and total lack of bias provided sound arguments in favor of trying out the infrared method during the IGY.

I also owe thanks to John Miller, present director of MLO, who suggested this article and allowed me time to complete it, and to Robert Bacastow, who devised the computer programs that executed many of the computations of this paper and who offered valuable criticisms. Financial support for the work described was by the Climate Dynamics Program of the U.S. National Science Foundation under grants ATM76-23053 and ATM77-25141.

REFERENCES

Arrhenius, S. A., 1903: Lehrbuch der Kosmischen Physik, V. 2, Hirzel, Leipzig, 477-481 Bacastow, R. B., 1976: Modulation of atmospheric carbon dioxide by the southern oscillation. Nature, 261:116-118. Bacastow, R. B., 1977: Influence of the southern oscillation on atmospheric carbon dioxide. In Fate of fossil fuel CO₂ in the oceans. Edited by N. R. Andersen and A. Malahoff, Plenum, N.Y., 33-43. Bischof, W., 1960: Periodical variations of the atmospheric CO2 content in Scandinavia. Tellus, 12:216-226. Bolin, B., and C. D. Keeling, 1963: Large-scale atmospheric mixing as deduced from the seasonal and meridional variations of carbon dioxide, J. Geophys. Res., 68:3899-3920. Bray, J. R., 1959: An analysis of the possible recent change in atmospheric carbon dioxide concentration, Tellus, 11:220-230. Buch, K., 1948: Der Kohlendioxydgehalt der Luft als Indikator der meteorologischen Luftqualität. Geophysica (Helsinki), 3:63-79. Callendar, G. S., 1938: The artificial production of carbon dioxide and its influence on temperature. Q. J. R. Meteorol. Soc. (London), 64:223-240. Callendar, G. S., 1940: Variations of the amount of carbon dioxide in different air currents, O. J. R. Meteorol. Soc. (London), 66:395-400.

Callendar, G. S., 1958: On the amount of carbon dioxide in the atmosphere. *Tellus*, 10:243-248.

- Eriksson, E., 1954: Report on an informal conference in atmospheric chemistry held at the Meteorological Institute, University of Stockholm, May 24-26, 1954. *Tellus*, 6:302-307.
- Fonselius, S., F. Koroleff, and K. Buch, 1955: Microdetermination of CO₂ in the air, with current data for Scandinavia. *Tellus*, 7:258-265.
- Fonselius, S., F. Koroleff, and K. E. Wärme, 1956: Carbon dioxide variations in the atmosphere. *Tellus*, 8:176-183.

Geophysics Study Committee, 1977: Overview and recommendations. In *Energy and climate*, National Academy of Sciences, Washington, D.C., 1-31.

- Hall, C. A. S., C. A. Ekdahl, and D. E. Wartenberg, 1975: A fifteen-year record of biotic metabolism in the Northern Hemisphere. *Nature*, 255:136-138.
- Keeling, C. D., 1958: The concentration and isotopic abundances of atmospheric carbon dioxide in rural areas. *Geochim. Cosmochim. Acta*, 13:322-334.

Keeling, C. D., 1960: The concentration and isotopic abundances of carbon dioxide in the atmosphere. *Tellus*, 12:200–203.

Keeling, C. D., 1961: The concentration and isotopic abundances of carbon dioxide in rural and marine air. *Geochim. Cosmochim. Acta*, 24:277-298. Keeling, C. D., R. B. Bacastow, A. E. Bainbridge, C. A. Ekdahl, P. R. Guenther, L. S. Waterman, and J. F. S. Chin, 1976a: Atmospheric carbon dioxide variations at Mauna Loa Observatory, Hawaii. *Tellus*, 28:538-551.

- Keeling, C. D., J. A. Adams, C. A. Ekdahl, and P. R. Guenther, 1976b: Atmospheric carbon dioxide variations at the South Pole. *Tellus*, 28:552-564.
- Keeling, C. D., and R. B. Bacastow, 1977: Impact of industrial gases on climate. In *Energy and climate*, National Academy of Sciences, Washington, D.C., 72-95.
- Machta, L., K. Hanson, and C. D. Keeling, 1976: Atmospheric carbon dioxide and some interpretations. In *Fate of fossil fuel CO₂ in* the oceans, Edited by N. R. Andersen and A. Malahoff, Plenum, N.Y., 131-144.
- Newell, R. E., and B. C. Weare, 1977: A relationship between atmospheric carbon dioxide and Pacific sea surface temperature. *Geophys. Res. Lett.*, 4:1–2.
- Reinsch, C. H., 1967: Smoothing by spline functions. Numerische Mathematik, 10:177-183.
- Revelle, R., and H. E. Suess, 1957: Carbon dioxide exchange between atmosphere and ocean, and the question of an increase of atmospheric CO₂ during the past decades. *Tellus*, 9:18–27.