The Concentration of Atmospheric Carbon Dioxide in Hawaii[1]

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Abstract. The concentration of atmospheric carbon dioxide at and near Mauna Loa Observatory, Hawaii, is reported for the first six years (1958–1963) of a long-term program to document the effects of the combustion of coal and petroleum on the distribution of CO_2 gas in the atmosphere and in the world's oceans. The majority of the measurements reported here were obtained at Mauna Loa Observatory with a continuously recording infrared gas analyzer. Also reported are measurements of 261 discrete samples of air collected in glass flasks on Mauna Loa, on the windward coast of Hawaii, and from aircraft near the Hawaiian Islands. The following results have been obtained: (1) The CO_2 concentration at Mauna Loa Observatory varies with season with an average amplitude of 6 ppm and is increasing at the average rate of 0.7 ppm per year. (2) These variations reflect regional changes in the air which lies above the trade wind layer near Hawaii. (3) The concentration of CO_2 in the trade wind layer is essentially the same as aloft except during the summer months when it is slightly lower. (4) Volcanic emanations of CO_2 near the summit of Mauna Loa and uptake of CO_2 on the forested lower slopes of the mountain influence the concentration of CO_2 at Mauna Loa Observatory but do not seriously interfere with the determination of regional changes.

INTRODUCTION

The great industrial demands for energy since the beginning of the 20th century have caused vast quantities of carbon dioxide to be released into the earth's atmosphere as a by-product of the burning of fossil fuels. The oceans are ultimately capable of absorbing most of this increase but, according to indirect estimates, cannot keep pace with the release [*Revelle and Suess,* 1957; *Bolin and Eriksson,* 1959]. Direct measurements of atmospheric carbon dioxide reported since late in the 19th century tend to show an increase in concentration [*Callendar,* 1958], but some question exists whether the observed trend is statistically significant [*Slocum,* 1955; *Bray,* 1959]. A revival by *Plass* [1956] and others of the hypothesis of *Tyndall* [1861] that the earth's climate is strongly affected by variations in the amount of atmospheric carbon dioxide justifies settling the question of the true rate of increase through a series of accurate measurements, either high in the atmosphere or on the earth's surface at locations well removed from local disturbing influences.

The International Geophysical Year provided an opportunity to try both these approaches, and in 1956 an extensive project to monitor atmospheric carbon dioxide was organized [*Fritz et al.,* 1959]. In 1958 accurate continuous surface measurements began in Antarctica and at Mauna Loa Observatory, then a newly established U. S. Weather Bureau meteorological research station on the island of Hawaii. Also, samples of air were collected in glass flasks 3000 to 6000 m above the North Pacific Ocean during weather reconnaissance flights of the United States Air Force. In 1960 collections in glass flasks began in Hawaii and from ships in the Pacific and Indian oceans. In 1961 continuous surface measurements began at Point Barrow, Alaska [*Kelley and Church,* 1962]. Most phases of the program continued until 1962 or 1963.

This article reports in detail the results obtained from Mauna Loa Observatory. A com-

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panion article [*Brown and Keeling*, 1965] reports results from Antarctica. Similar reports of the aircraft, ship, and Point Barrow projects are in preparation. A general summary of all of the data up to April 1962 has been presented by *Bolin and Keeling* [1963].

Observing Site

Mauna Loa Observatory is located on the north slope of the volcano Mauna Loa (Figure 1). The observatory is 3400 m above sea level and 800 m below the summit of the mountain. It is situated at 19.5°N, 155.6°W, well within the geographic tropics, south and west of the semipermanent eastern Pacific atmospheric high-pressure cell. The prevailing surface wind in the vicinity of the island is the northeasterly trade wind, which prevails 80% of the year. Over the ocean this steady moist wind extends upward from the surface to a temperature inversion with a mean height of 2000 m. Still higher, the air is usually dry and wind speeds are low, but on Mauna Loa heating of the mountain slopes by the sun during the day causes moist air from below the inversion to seep up the mountain, often to 3000 m or higher, so that air having some of the characteristics of the trade wind frequently envelopes the observatory in the afternoon. At night the slopes of the mountain cool, a dry descending breeze is generated, and the moist air recedes to a level near or below the height of the oceanic trade wind inversion. Only occasionally does this diurnal regime disappear—for example, during those relatively rare periods when strong winds develop above the trade winds. Many properties of the air at the observatory, therefore, show marked diurnal variability. But since the

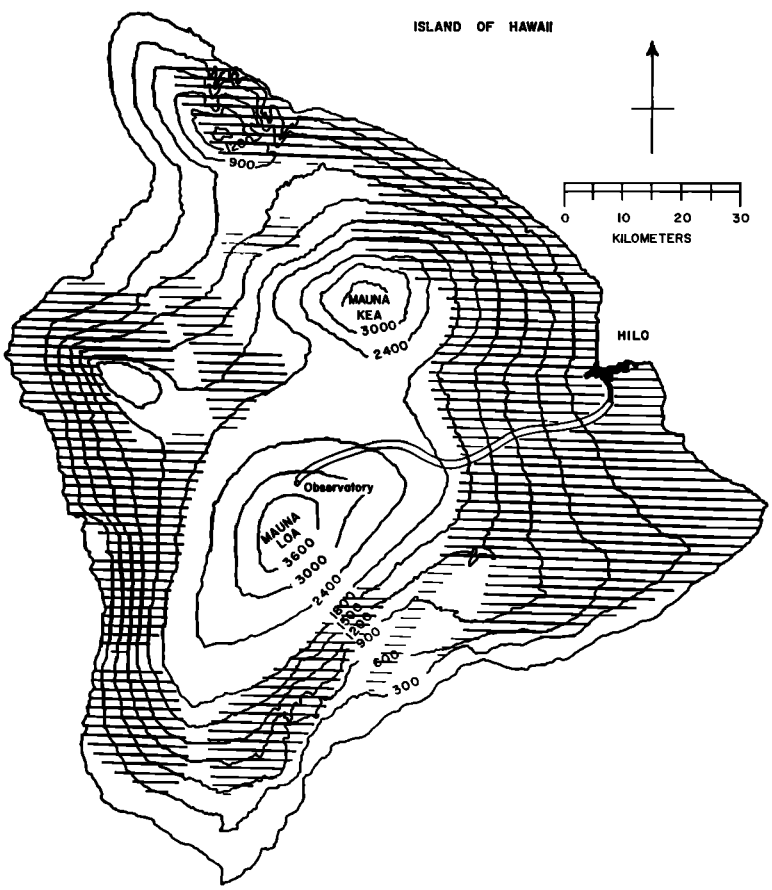


Fig. 1. Contour map of Hawaii showing the location of Mauna Loa Observatory. Figures are heights above sea level in meters. The horizontal shading indicates wooded areas.

air which descends the upper slopes of Mauna Loa at night is largely derived from air aloft, a basis is available for relating the complicated features of the daytime air to air undisturbed by the local regime.

Mauna Loa Observatory and its meteorological milieu have been described by *Price and Pales* [1959]. A general description of the climate of the Hawaiian Islands has been compiled by *Blumenstock* [1961].

The observatory is remote from sources and sinks of carbon dioxide except for sources at the station itself. Careful placing of intakes for sampling the air prevents serious contamination. The nearest continental land mass is too far away (3700 km) to have any regional influence. The nearest vegetation of consequence is 30 km from the observatory and below the prevailing trade wind inversion. The influence of plant activity is often perceptible during the latter half of the day during periods of upslope winds, but it is otherwise not detectable. Outgassing from volcanic vents upslope from the observatory affects the air on some nights, but its influence on the CO_2 concentration of the air is readily discerned and can be eliminated from consideration. The observatory is thus an excellent, if not an ideal, site for measuring CO_2 in the upper air of the northern tropics.

Experimental Procedures

The gas analyzer. Continuous measurements of atmospheric CO_2 relative to dry air have been made with a nondispersive infrared ana-

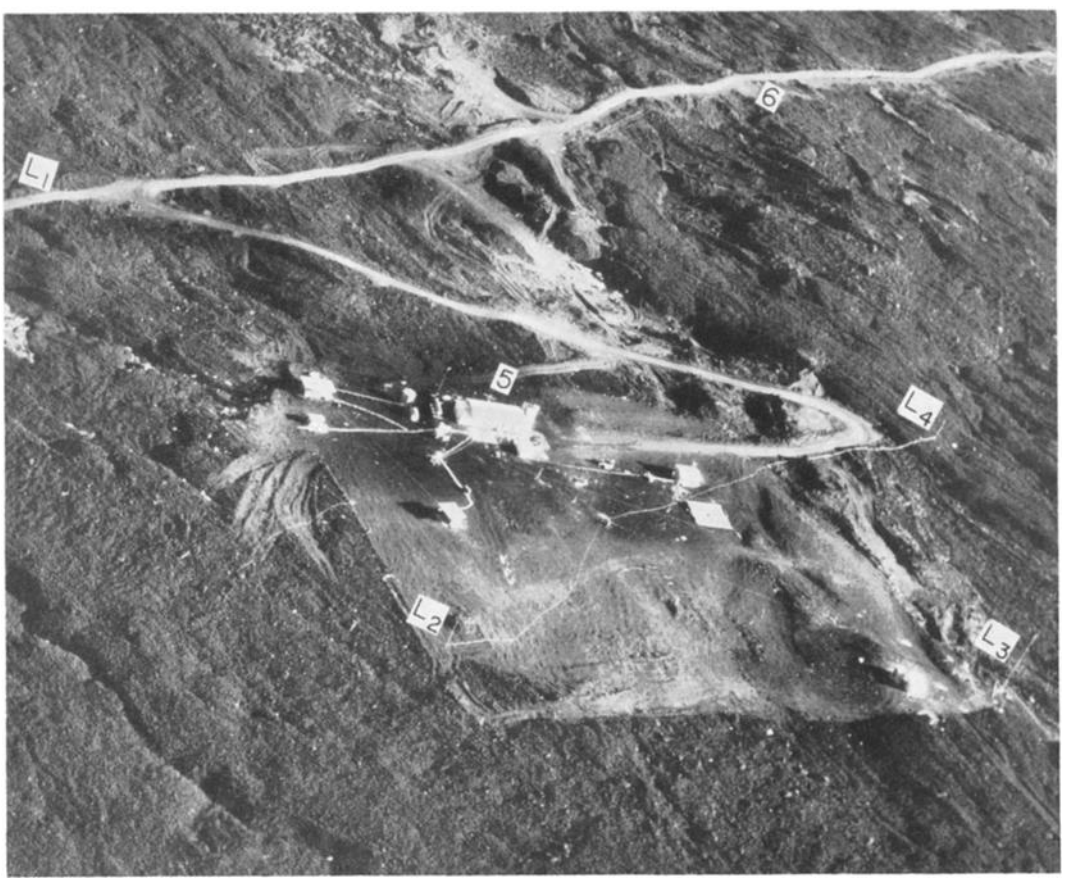


Fig. 2. Aerial view of the observatory, looking north (downslope). L_1 to L_4 are air intake towers. (5) is the main observatory building which houses the analyzer. Aluminum tubing between the towers and the observatory is partially visible. (6) is the road to Hilo. Photograph was taken in August 1962.

lyzer specifically designed for the monitoring of flowing air. This instrument, described by *Smith* [1953], measures the loss of energy of a beam of infrared radiation traversing a gas sample. The radiation, emitted by a glowing nichrome filament, is mechanically chopped at 20 cps and then directed through the gas sample into a detector cell which is permanently filled with CO_2 diluted with argon. Absorption of radiation by the CO_2 gas in the detector cell produces a cyclic pulsation in pressure which is transmitted to a tantalum diaphragm of a condenser microphone and thence converted to an alternating voltage, amplified, and recorded. The presence of CO_2 in the gas sample stream reduces the radiation reaching the detector cell at just those wavelengths at which absorption can occur in the detector. Thus the voltage developed by the detector varies inversely with the concentration of CO_2 in the gas sample. This voltage is plotted by a strip chart recorder which thereby furnishes a continuous record of the concentration of CO_2 in the gas sample stream.

The air system. Four air intakes are mounted 7 m above ground level, 175 m distant, and in mutually perpendicular directions from the observatory's main building (Figure 2). Air lines of aluminum tubing with an inside diameter of 8 mm convey sample air to this building. Here the air lines connect to a selecting system, shown schematically in Figure 3. Taking one air line as an example, sample air enters the selecting system at E_2, at a flow rate of 5 to 10 l/min, drawn by a diaphragm pump P. A vacuum gage F registers the pressure in the air line, and a pressure gage L registers the pressure which the pump develops. A needle valve K reduces the air flow to 0.5 l/min. The excess air is exhausted into the room by a pressure relief valve R. The remaining air passes through a magnetic solenoid valve N_2, which can be energized either manually or automatically. When N_2 is energized as shown in Figure 3, air passes through the 'normally closed' outlet, then through a nonenergized solenoid valve B, and into the analyzer. When solenoid N_2 is not energized, air is diverted into the room through the 'normally open' outlet. When solenoid B is energized, but N_1 to N_4 are not, the flow of air to the analyzer is stopped, and reference gas

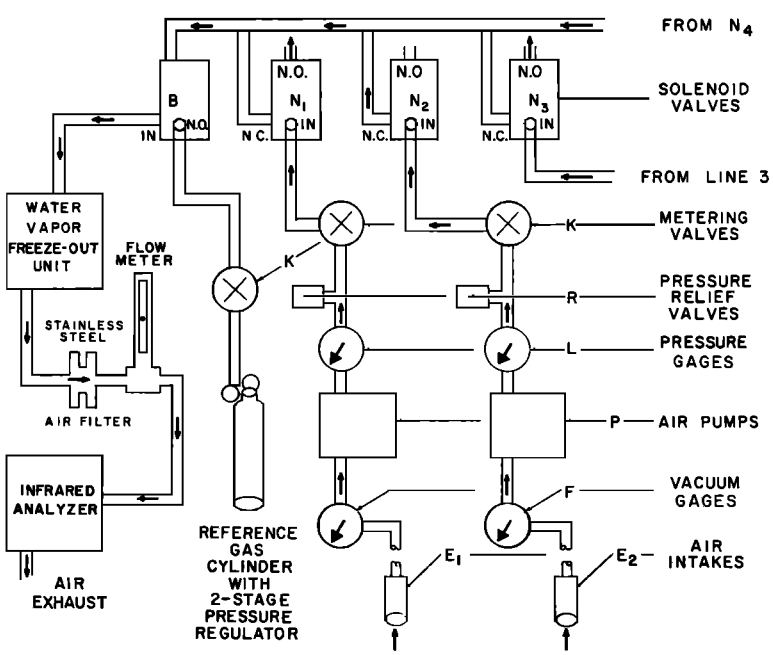


Fig. 3. Air flow system for atmospheric CO_2. Letters are explained in the text. N.O., normally open; N.C., normally closed.

CARBON DIOXIDE DATA STATION MAUNA LOA OBSERVATORY, HAWAII Sheet No. 780

Span 485 Zero 700 Freezer Temp. −83° Date JAN 5, 1961

Flow 0.6 lpm Working Ref. Tank 3758

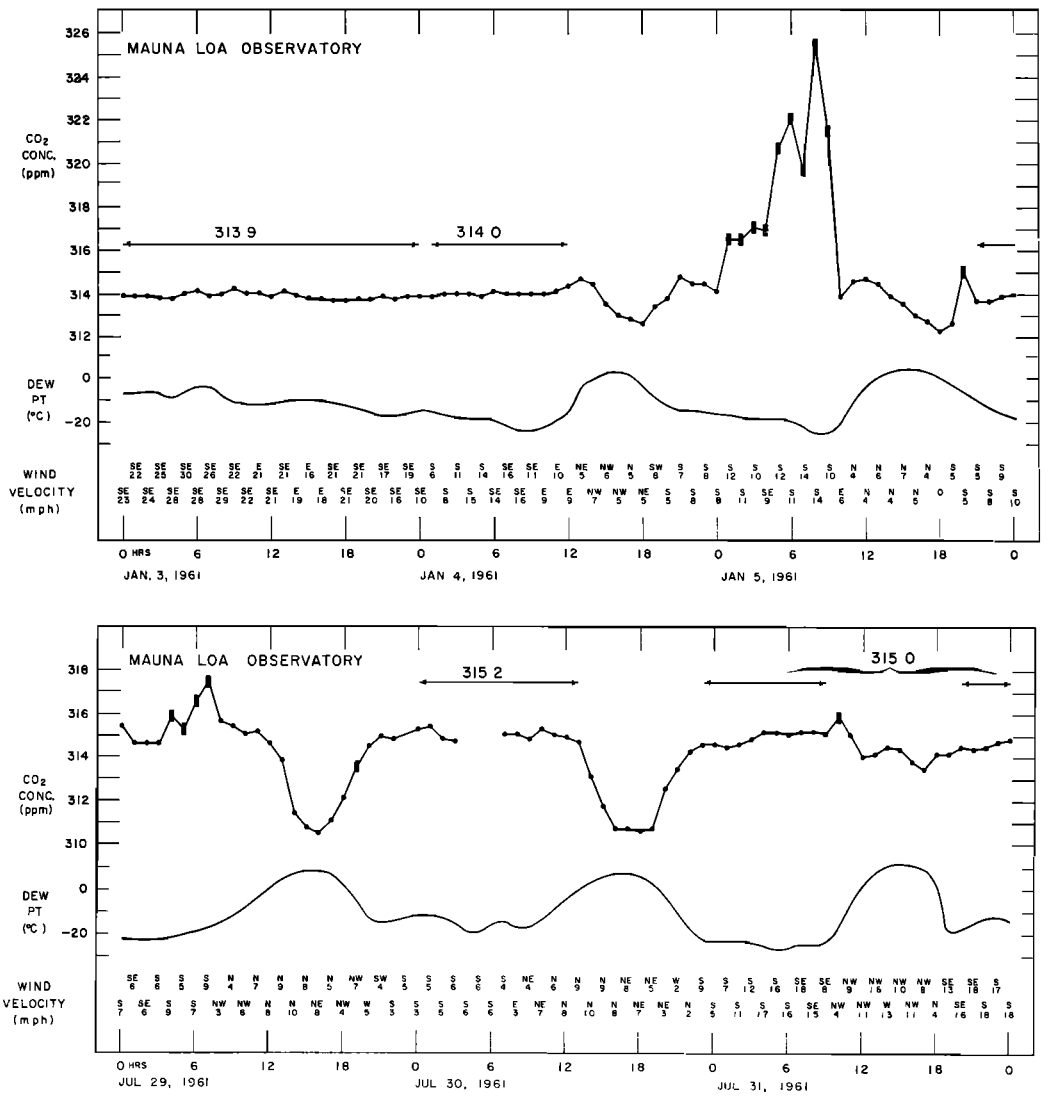
Tank Press. 1340 psi Obs. H.A.

	L1	L2	L3	L4	L Diff.	L Ave.	Sky	Vis (mi)	Weather	Sta. Press (In./Hg)	Temp (°F)	D.P. (°F)	Wind Dir	Wind Vel (MPH)	R.H.	Wet Bulb (°F)	REMARKS
0030		5.0	3.4		1.5					20							
0100		4.5	8.0		3.5	5.2				070	34	1	S	12	25		
0130		4.8	4.8														
0200		7.5	3.7		3.6	5.2				060	34	1	S	11	24		
0230		4.2	7.6		3.3												
0300		6.9	4.1		2.3	5.7				055	33	−1	S	10	22		
0330		4.8	4.3		0.5												
0400		5.9	7.4		1.8	5.6				055	34	−1	SE	9	23		
0430		6.3	6.3														
0500		11.0	11.3			8.9				056	34	−1	S	12	22		
0530		9.0	8.6														
0600		11.5	11.3			10.1				055	33	−2	S	11	21		
0630		8.5	6.8		1.7												
0700		8.1	8.7		6.9	8.0	O	80		.055	32.8	−8	S	14	17	21.2	
0730		8.1	7.8														
0800		23.8	12.5		11.0	13.1	O	80		.065	37.0	−6	S	14	16	23.5	
0830		16.0	13.0		3.0												
0900		6.0	3.2		2.4	9.6	O	80		.070	46.8	−19	S	10	6	28.1	
0930			3.1	3.1													
1000			2.6	2.7		2.9	O	80		.070	47.1	−3	E	6	12	29.7	
1030			3.2	2.6	0.7												
1100			4.3	3.8		3.5	O	80		.080	46.8	14	N	4	28	32.9	
1130			3.8	3.6	0.3												
1200	3.4			3.7		3.6	O	80		.070	46.6	21	N	4	36	34.9	
1230	3.3			3.5													
1300	3.2			3.6		3.4	O	80		.055	46.2	36	N	6	66	40.4	
1330	3.0			2.8													
1400	3.0			2.9		2.9	O	80		.025	48.1	35	N	4	60	41.2	
1430	2.6			2.6													
1500	2.7			2.3		2.6	E 30 ⓓ	40		.025	44.0	40	N	7	85	41.5	
1530	2.3			2.1													
1600	2.2			1.9		2.1	E 30 ⓓ	80		.025	43.5	39	N	5	85	41.2	
1630	1.9			1.6													
1700	2.1			1.8		1.9	O	80		.025	44.2	40	N	4	84	41.7	
1730	1.4			1.7													
1800	1.5			1.5	0.5	1.5				.030	44	20		C	98		
1830	1.8			1.4	0.7												
1900	1.7		2.1			1.8				.035	42	24	S	5	48		
1930		2.4	2.7		1.0												
2000		4.8	4.8			3.9				.040	39	20	S	5	48		
2030		2.6	2.6														
2100		2.6	2.8			2.7				.045	37	14	S	5	38		
2130		2.8	2.8														
2200		2.7	3.5		1.0	2.7				.045	36	8	S	8	30		
2230		2.8	2.8														
2300		2.9	3.0			2.9				.045	36	4	S	9	24		
2330		2.9	3.0														
2400		3.0	3.1			3.0				.040	36	0	S	10	22		

HH

Fig. 4. A daily data sheet showing original values obtained by the infrared analyzer for CO_2, plus hourly values of meteorological variables. Some of the data listed here are also plotted in Figure 5*a*.

flows into the analyzer through *B*. A freeze-out trap maintained at approximately −80°C removes water vapor which, because of infrared spectral bands which overlap those of CO_2, would otherwise interfere with the analysis. A stainless steel air filter protects the analyzer cell from dust and a flow meter registers the flow. The sequence of sampling is controlled electrically by a timer and plug board selector which allows automatic sampling from a preselected



Figs. 5*a* and 5*b*. An illustration of the hourly average concentration of atmospheric CO_2 at Mauna Loa Observatory versus local time. Values are shown by connected points or vertical bars. The latter denote periods in which the recorder's pen trace was variable. Horizontal arrows above the plot indicate periods of steady concentration, usually divided into calendar days. The average concentration for each steady period is indicated, except for January 5 when the period was so short that it was included with the following day (not shown). The dew point as measured by a continuously recording infrared hygrometer is shown by a smoothed curve. Values are given of the hourly average wind velocity from a recording cup anemometer. Wind speed is in miles per hour (m/sec × 2.238).

air intake for 10 min, from another air intake for 10 min, and then from reference gas for 10 min. The two intakes selected for sampling are those most nearly upwind from the station.

Reference gas. For calibration of the analyzer, mixtures of CO_2 in nitrogen of high purity are prepared with a CO_2 concentration in the same range as atmospheric air. These 'reference gases' are stored in size 3*A* stainless steel cylinders at an initial pressure of 150 atm. A cylinder holds approximately 6000 standard liters of gas. Normally four reference gases are used together. Three are semipermanent calibrating standards whose concentrations are determined at Scripps before and after use. The fourth is a 'working reference,' which is compared every 30 min with air. Each working reference gas receives a close check of its concentration relative to the three calibrating standards throughout its period of use, plus checks at Scripps before and after use. All comparisons are used to determine the average concentration of each working gas. This final value is accurate to approximately ±0.1 ppm.

Logging the data. Normally, four comparisons of air versus reference gas are obtained each hour and registered on the recorder chart. The difference, in recorder chart ordinates, between successive traces is read where the trace registers a change between air and reference gas. These differences are entered on daily data sheets (Figure 4). If a 10-min trace for air shows variations of greater amplitude than are shown by the nearest trace for reference gas, the comparison based on these traces is marked *v* for 'variable'. Data pertaining to each air line are entered in a separate column to aid in detecting any systematic difference between air lines, which would be a sign of contamination. Any directly observable differences between traces for adjacent air lines are recorded in a separate column as a direct proof of contamination.

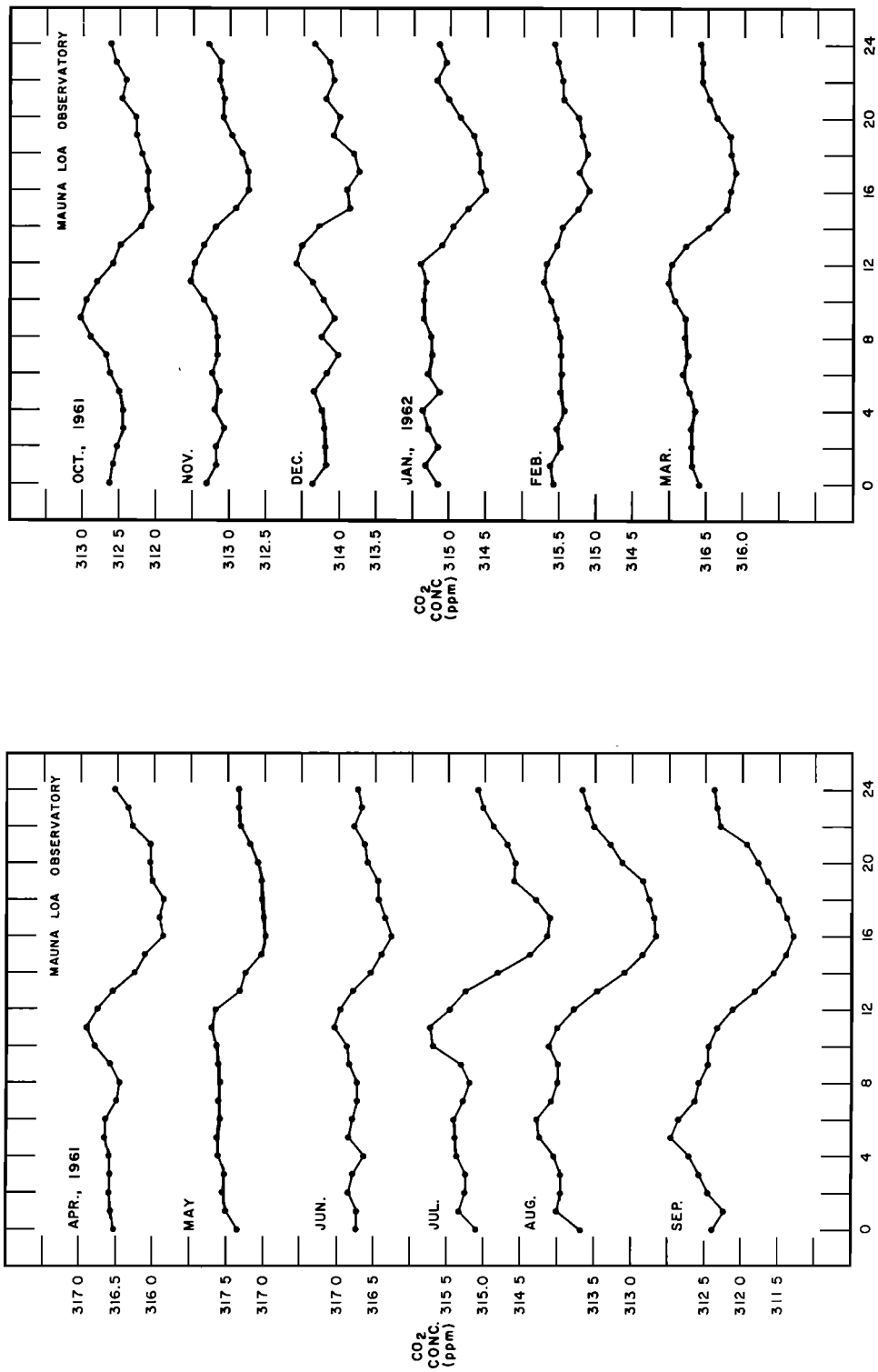
From the recorded data a time plot is made of the hourly average difference in chart ordinates between air and reference gas. Two excerpts of this plot are shown in Figures 5*a* and 5*b*. These figures show the variability often found in the CO_2 concentration at Mauna Loa. To obtain average values which reflect the prevailing concentration over the adjacent Pacific Ocean, we average those portions of the record in which the concentration has been steady for 6 hours or more. We reject 'variable' comparisons which fall within steady periods. To illustrate this, the portions selected for averaging for January 3 to 5 and July 29 to 31, 1961, are shown in Figures 5*a* and 5*b*. Daily averages of the ordinate differences are computed from the selected portions of the record. Daily concentrations of CO_2 in parts per million of dry air by volume (ppm) are calculated from these on the basis of the preliminary calibrations of the reference gases available at that time.

All the above operations are completed with the least possible delay so that evidence of contamination or other unusual behavior of the observed CO_2 concentration at the station can be quickly noted. Final values of the concentration are obtained only after all of the reference gases have received final analyses at Scripps—a delay of up to 2 years. The preliminary and final values, however, usually differ by less than the experimental uncertainty in the air data (about ±0.2 ppm).

Flask sampling. Since March 1960, samples of air have been collected in 5-liter glass flasks at Mauna Loa Observatory and at sea level on the east-facing windward coast of Hawaii near Hilo. The flasks, previously pumped to a high vacuum, are exposed by opening a greased stopcock so that air expands into the flask. The samples are returned to Scripps for laboratory analysis. The accuracy of individual analyses is approximately ±0.3 ppm.

During 1960 and 1961, samples were also collected from aircraft at the 700-mb pressure level on weather reconnaissance flights of the U. S. Air Force. The flights, bearing the Air Force name 'Loon Kilo,' originated near Honolulu, Hawaii, and covered a region within approximately 1500 km of Mauna Loa. These samples were collected in 1.8-liter glass flasks. Experimental details will be given in a separate paper dealing specifically with the aircraft project.

Supplementary data. The individual reference gas calibrations and atmospheric air measurements are too extensive to reproduce in this article. They have been summarized in a detailed report [*Keeling and Pales,* 1965] and deposited with the American Documentation Institute. This report includes all relevant calculations and supporting data. Final daily aver-



Figs. 6*a* and 6*b*. The diurnal course of atmospheric CO_2 at Mauna Loa Observatory.

age values of the CO_2 concentration are tabulated.[3]

Continuous Data

The concentration of atmospheric CO_2 at Mauna Loa Observatory as measured by the gas analyzer varies over a wide range of time scales. Concentrations are reported in parts per million of dry air (ppm).

Short-term changes. Two types of short-term variability occur frequently at Mauna Loa: (1) irregular bursts of high concentration which occur during about 20% of all nights and (2) gradual dips in concentration which occur during about 60% of all afternoons. Both phenomena are illustrated in Figures 5*a* and 5*b*. The nocturnal bursts invariably appear as irregular traces on the original recorder charts, a feature which suggests imperfect mixing of CO_2 injected into the atmosphere nearby. The afternoon dips are less irregular, which suggests a widespread sink at greater distance. The traces in the afternoon are often smooth even during periods when the concentration is rising or falling sharply.

Afternoon dips occur with high persistence. They influence the average diurnal course for every month of the year, as is seen by plotting hourly the monthly average concentration, based on all traces except those noted 'variable,' from April 1961 to March 1962 (Figures 6*a* and 6*b*). Omitting the variable traces removed the influence of the nocturnal bursts but did not cancel the afternoon dips.

The monthly average amplitude of the dip varies. This is seen by plotting the difference between the average forenoon concentration and the lowest value during the afternoon dip (Figure 7). The amplitude is exceptionally large during July, August, and September.

The monthly plots of the diurnal course are somewhat irregular during the forenoon, but a plot of the yearly average diurnal course shows no significant variation except for the clearly evident dip during the afternoon (Figure 8).

If we consider these features and the diurnal regime at the observatory as discussed above, the evidence suggests that the dips are principally a result of CO_2 uptake by island vegetation on the lower slopes of Mauna Loa. The exceptionally large dips observed during July, August, and September probably reflect a depletion of CO_2 in the air of the trade winds upwind of the island relative to air above the trade wind inversion, as observed by *Bolin and Keeling* [1963].

Nocturnal bursts occur less frequently than afternoon dips and are essentially random (Figures 9*a* to 9*f*). The average frequency of nights with bursts is 22%, and does not vary significantly from year to year. The frequency during the summer months is slightly less than at other seasons, but the difference is insignificant.

A striking example of a nocturnal burst is seen in the plot for January 4–5 (Figure 5*a*). Two general characteristics of nocturnal bursts are well illustrated in this example: (1) a similarity of wind speed and direction with the previous night in which no trace of a burst can be seen and (2) termination of the burst when the wind shifted from south to east. These features are consistent with the general finding that a southerly wind is a necessary, but not sufficient, condition for bursts. Considering this evidence and that they occur randomly, we believe that the bursts are produced by intermittent sources of volcanic CO_2 from near the summit of the mountain.

Both the afternoon dips and nocturnal bursts diminish in intensity with increasing wind speed. When the average wind speed exceeds about 10 m/sec, the hourly values are usually steady, as for January 3, 1961 (Figure 5*a*). Steady concentrations may occur at night even with low wind speeds, but during the day the concentration rarely remains steady in the absence of strong winds. The steady concentration during January 3 persisted through the night after the wind abated, but by 1200 on January 4 the typical diurnal pattern had returned. This is confirmed by the shift in wind to the northern quadrant and by the large increase in humidity in the afternoon, in contrast to the previous day's steady wind and low, slowly varying humidity.

[3] This information has been deposited as Document 8530 with the ADI Auxiliary Publications Project, Photoduplication Service, Library of Congress, Washington, D. C. A copy may be obtained by citing the document number and remitting $22.50 for photoprints or $6.50 for 35-mm microfilm. Advance payment is required. Make check or money order payable to: Chief, Photoduplication Service, Library of Congress.

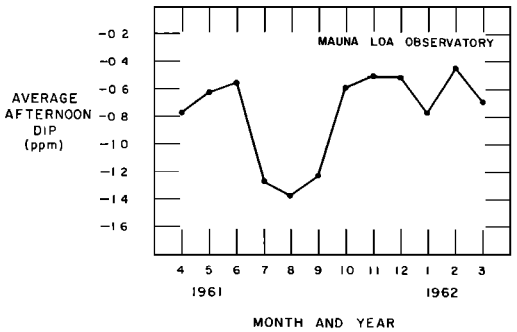


Fig. 7. The average intensity of the afternoon dip at Mauna Loa Observatory for the same months as shown in Figure 6.

The plot for July 29–31, 1961 (Figure 5*b*), illustrates a common regime in summer, with dry downslope winds at night and moist upslope winds during the day. One small nocturnal burst occurred during the 3-day period; afternoon dips occurred every day.

The slight peak at 1000 hrs on July 31 was associated with a wind shift from southeast to northwest which, for a brief time, caused nearly the same air to pass over the station a second time and thus brought local contamination to the upwind air intakes.

The seasonal course. Daily average concentrations are plotted in Figures 9*a* to 9*f*. The averages were computed for steady periods as described above. The nocturnal bursts are seen to have no special influence on the daily concentration of CO_2 except to reduce the probability that a daily concentration will have been computed.

A seasonal oscillation shows clearly in the plots. This feature is characteristic of air over the northern Pacific Ocean at all latitudes. A detailed study of amplitudes and phases at different latitudes [*Bolin and Keeling,* 1963], strongly supports a hypothesis that the oscillations are produced principally by land plants, especially forests of North America and Asia, which withdraw CO_2 from the atmosphere during summer and release CO_2 to the atmosphere during autumn, winter, and spring.

Shorter-term trends in CO_2 concentration, with a duration of 5 to 10 days, are occassionally discernible in Figures 9*a* to 9*f*. These trends suggest a minor association of CO_2 concentrations with regional weather patterns but are too small to permit us to draw conclusions.

Monthly averages of the daily average concentrations are listed in Table 1 and plotted in Figure 10. The seasonal oscillation repeats from year to year with an upward trend. The progression of monthly values is remarkably steady.

Long-term changes. The result of smoothing the data of Table 1 by using a 12-month running mean is shown in Figure 11. Fitting the means to a linear relation by the method of least squares yields the straight line shown in Figure 11. The slope is 0.68 ppm/yr. The standard deviation of individual means from the least-squares relation is 0.10 ppm. The rate of increase for Mauna Loa integrated over the entire atmosphere is approximately one-half the rate at which CO_2 is released into the atmosphere by the combustion of fossil fuels [*Revelle and Suess,* 1957].

Figure 11 suggests the possibility of a weak 2-year cycle, but the time record is too short to establish its significance.

Flask Data

The measurements discussed above have been supplemented with measurements of air collected in evacuated glass flasks and analyzed at Scripps. Collections at the observatory allow a check of the analyzer against the flask technique; collections on the windward coast near Hilo allow a comparison of air at Mauna Loa Observatory with air near sea level; collections from aircraft allow a comparison of surface air with regionally undisturbed air aloft.

Collections at Mauna Loa Observatory. Table 2 gives the concentration of CO_2 in individual flask samples collected at Mauna Loa Observatory, the concentration observed by the gas analyzer at the time of exposure, and the departures of the analyzer values from the flask values.

To minimize the influence of contamination, departures were computed only if two or more flasks, exposed a few minutes apart, yielded values which agreed to within 1.0 ppm. This was done on the supposition that, if samples had been contaminated either by the observer's breath or by combustion from the station, the level of contamination would usually vary from one flask to the next. The closest tolerance for rejection that could be set was 1.0 ppm, the approximate maximum error of analysis.

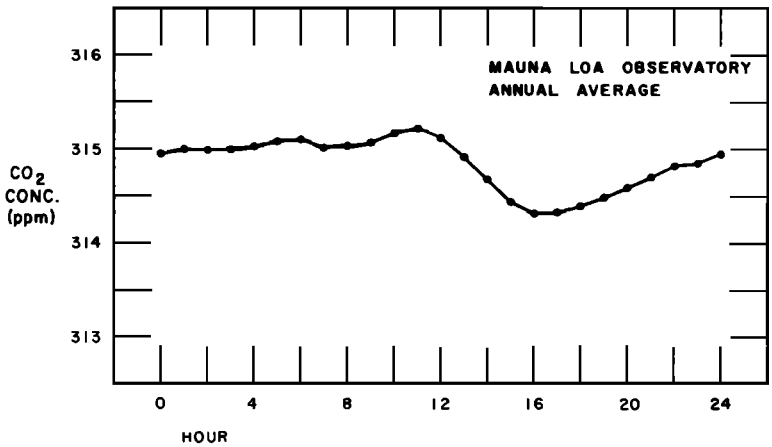


Fig. 8. The annual average diurnal course of atmospheric CO_2 at Mauna Loa Observatory for the same period as shown in Figure 6.

This criterion did not invariably eliminate serious contamination. On two occasions (August 1 and December 15, 1961) samples, taken downwind of the observatory by mistake, agreed closely with each other. Indeed, on December 15, the samples were contaminated by over 4 ppm but duplicates yielded identical values. Rejecting these two pairs, we consider the results reliable.

Measurements by the analyzer have been on an average 0.2 ppm lower than the flask measurements, if those flask samples judged to be contaminated are excluded.

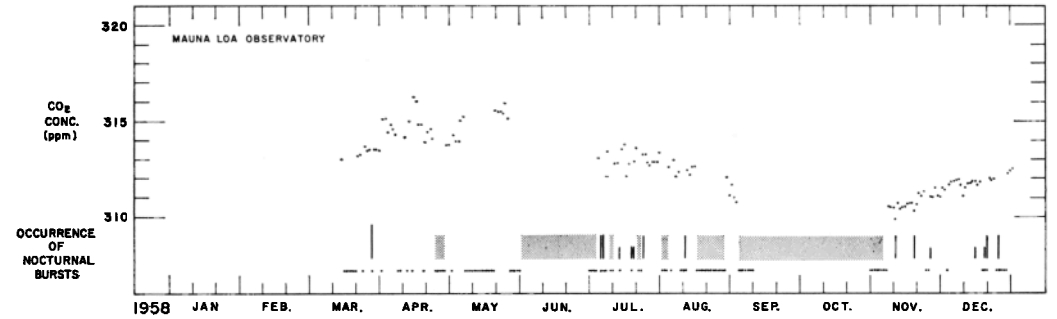
Collections near Hilo. The results of flask sampling in the vicinity of Hilo are given in Table 3. Flasks were exposed on a beach about 1 km east of the city. In most cases pairs of samples were taken. The concentrations found for the individual samples can be reconstructed from the average values and ranges given for each sampling period. The considerable scatter is evidence of some contamination. This is also suggested by the generally poorer agreement between duplicate samples, as compared with those exposed at Mauna Loa Observatory. During July, August, and September concentrations lower than at Mauna Loa were often observed. During other months the concentrations were nearly always higher.

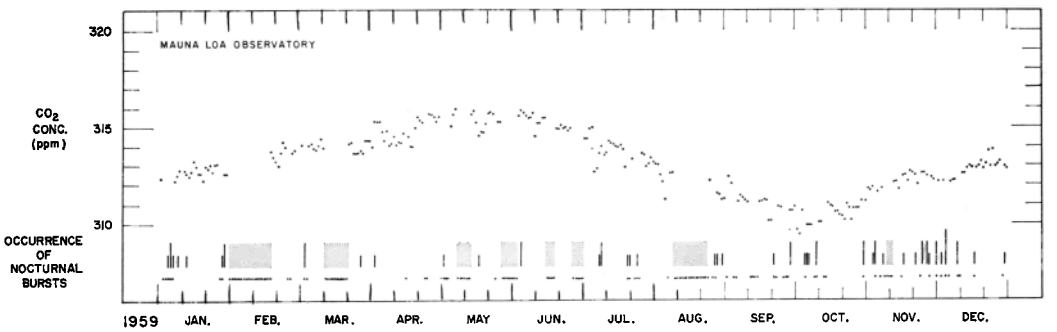
Collections from aircraft. Groups of approximately 12 samples each were obtained on seven weather reconnaissance flights during 1960 and 1961. Areal plots of the individual measurements are shown in Figures 12*a* to 12*g*. Figures 13*a* and 13*b* show two examples of the hourly average concentration for Mauna Loa during a 3-day period chosen to include the largest proportion of steady record to occur within 3 days of a flight.

In Table 4 the average concentrations for the steady periods at Mauna Loa nearest the time of aircraft sampling have been entered beside the averages of those aircraft data enclosed by dashed circles. Differences are shown in a third column. With one exception the agreement is satisfactory. The August data need further comment.

August is in the season when significant vertical and north-south gradients occur generally over the North Pacific Ocean with diminishing concentration downward and northward [*Bolin and Keeling,* 1963]. The record at Mauna Loa is generally most variable at this season; the afternoon dips are exceptionally large; the concentration near Hilo often is lower than that at the observatory. The only steady record falling within 3 days of the aircraft sampling occurred during the afternoon of August 2 (see Figure 13). That Mauna Loa Observatory registered an average concentration 2.0 ppm lower than the aircraft samples is probably a result of the advection of air from considerably below the 700-mb level.

If all flights except those in August are con-


MAUNA LOA OBSERVATORY
CO_2 CONC. (ppm)
320
315
310
OCCURRENCE OF NOCTURNAL BURSTS
1958 JAN FEB. MAR. APR. MAY JUN. JUL. AUG. SEP. OCT. NOV. DEC.


MAUNA LOA OBSERVATORY
CO_2 CONC. (ppm)
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OCCURRENCE OF NOCTURNAL BURSTS
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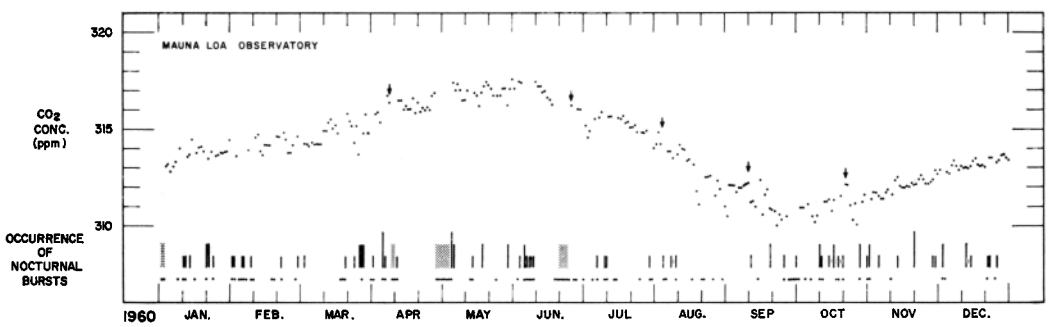
MAUNA LOA OBSERVATORY
CO_2 CONC. (ppm)
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OCCURRENCE OF NOCTURNAL BURSTS
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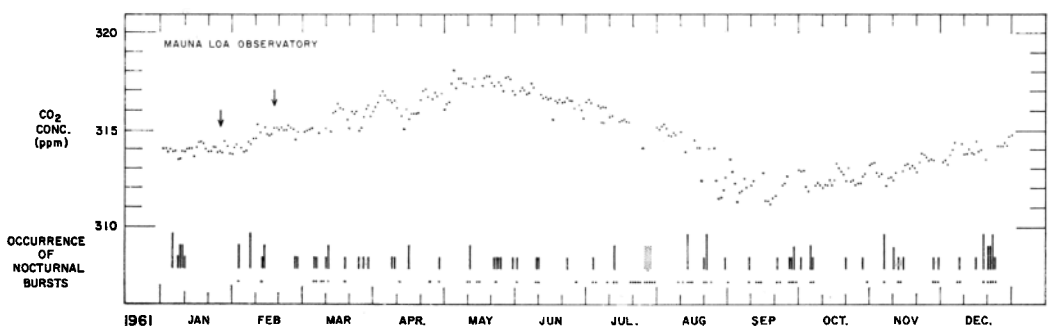
MAUNA LOA OBSERVATORY
CO_2 CONC. (ppm)
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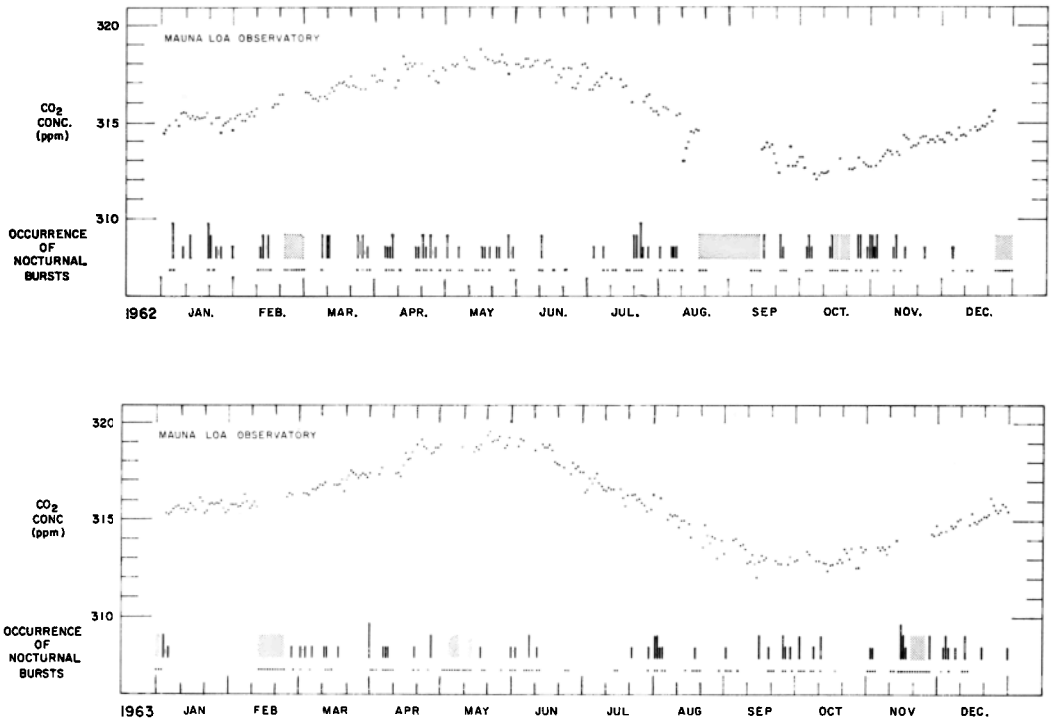
OCCURRENCE OF NOCTURNAL BURSTS
1961 JAN FEB MAR APR. MAY JUN JUL. AUG SEP OCT. NOV DEC.



Figs. 9*a* to 9*f*. The daily average concentration of atmospheric CO_2 versus time at Mauna Loa Observatory for periods of steady concentration. Values are shown by unconnected points. If no value was determined on a given day, a point appears below the main plot as a reading aid. Vertical lines denote days in which nocturnal bursts occurred within a 24-hour period commencing 12 hours before the calendar day. The approximate intensity of the maximum burst is indicated by the length of the line: shortest lines denote 0 to 5 ppm; medium-length lines, 5 to 10 ppm; longest lines, 10 to 15 ppm. Shaded areas represent periods when the analyzer was shut down for 48 hours or more. Small vertical arrows during 1960 and 1961 mark days of aircraft sampling as shown in Figures 12*a* to 12*g*.

sidered, the steady record at Mauna Loa yields data which average 0.2 ppm lower than regionally undisturbed air.

Discussion

Representativeness of the data. In assessing the Mauna Loa measurements, our primary objective is to establish how well they represent atmospheric CO_2 uninfluenced by the Hawaiian Islands. Some of the measurements are clearly influenced locally, as seen by significant short-term fluctuations in concentration such as nearby sources and sinks produce. But how free are the remaining data, based on periods of steady concentration, of local disturbances?

The omission of variable periods from the daily averages eliminates almost all influences of the nocturnal 'bursts' that occasionally disturb the record at night, but it does not eliminate the 'dips' that occur frequently in the afternoon. The extent to which these dips influence the record can be checked, however. In Table 5 the monthly averages for steady periods are compared with the monthly averages for 0–12 hours and for 0–24 hours of all traces except those initially noted to be 'variable,' from April 1961 through March 1962. The steady data agree more closely with the 0–24 hour averages, but differences are slight. The inclusion of the afternoon data reduces the average yearly concentration from 315.07 to 314.85 ppm. Thus afternoon dips have lowered the averages for steady periods by only about 0.2 ppm.

TABLE 1. Monthly Average Concentration of Atmospheric Carbon Dioxide (ppm) at Mauna Loa Observatory

Month	No. of Days	Average Concentration	No. of Days	Average Concentration	No. of Days	Average Concentration	No. of Days	Average Concentration	No. of Days	Average Concentration	No. of Days	Average Concentration
	1958		1959		1960		1961		1962		1963	
January			22	312.75	23	313.67	31	313.96	27	314.97	27	315.63
February			9	313.70	15	314.20	26	314.71	15	315.48	15	315.90
March	9	313.50	15	314.04	22	314.92	25	315.58	25	316.60	26	316.94
April	15	314.73	26	314.85	22	316.20	27	316.36	19	317.47	19	318.25
May	9	315.06	15	315.35	23	316.99	26	317.37	20	318.03	14	318.98
June			17	315.30	13	316.71	26	316.62	26	317.65	19	318.25
July	17	313.08	25	313.75	19	315.15	16	315.70	20	316.40	27	316.46
August	9	312.31	9	312.06	21	313.03	22	313.80	12	314.63	21	314.43
September	3	311.19	16	311.23	21	311.28	20	312.14	14	313.17	20	313.09
October			23	310.69	19	311.11	28	312.53	19	312.52	23	313.01
November	18	310.80	18	312.23	28	312.07	24	313.09	26	313.72	10	313.83
December	22	311.86	23	312.84	26	313.20	24	314.06	19	314.56	26	315.15
Annual Mean		312.82		313.23		314 04		314.66		315.43		315.83

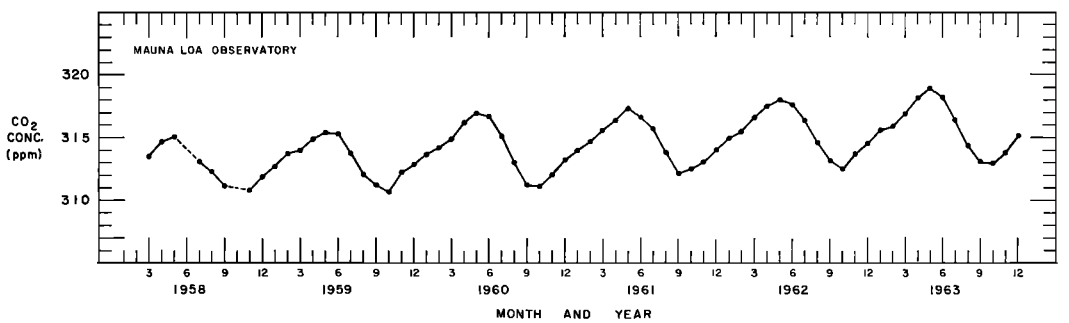


Fig. 10. Monthly average concentration of atmospheric CO_2 at Mauna Loa Observatory versus time.

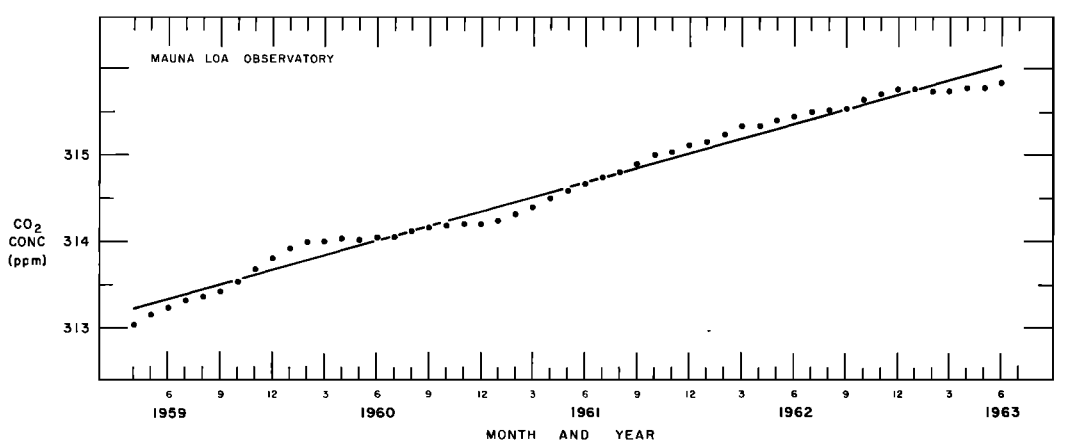


Fig. 11. Twelve-month running mean of the concentration of atmospheric CO_2 at Mauna Loa Observatory. Means are plotted versus the sixth month of the appropriate 12-month interval. The straight line indicates a rate of increase of 0.68 ppm/yr.

TABLE 2. Comparisons of Analyzer and Flask Results at Mauna Loa Observatory

Date	Local Time	CO_2 Concentration, ppm: Flask	CO_2 Concentration, ppm: Analyzer	Departure,* ppm
1960				
March 31	1600	316.3	315.8	−0.5
		315.8		0.0
		316.5		−0.7
April 23	1030	316.6	316.0	−0.6
		316.8		−0.8
		316.5		−0.5
April 24	0800	317.1	316.1	−1.0
		316.9		−0.8
		317.2		−1.1
July 3	1520	315.0	314.8	
		321.0		
July 17	1700	314.1	313 9	−0.2
		313.2		0.7
Aug. 1	1500	312.4	312.0	
		321.6		
Aug. 18	2000	313.5	312.9	−0.6
		313.6		−0.7
Oct. 6	1450	310.4	309.8	−0.6
		310.4		−0.6
Nov. 4	1345	315.8	311.6	
		374.7		
Dec. 1	1050	313.6	312.6	−1.0
		313.6		−1.0
1961				
Jan. 4	1250	314.0	314.7	0.7
		314.0		0.7
Jan. 17	1400	315.1	314.4	−0.7
		315.1		−0.7
Feb. 2	1350	315.2	313.8	−1.4
		314.6		−0.8
Feb. 17	1115	316.1	315.4	−0.7
		315.5		−0.1
March 14	1520	314.8	315.6	
April 4	1110	318.6	317.5	
		317.2		
May 15	1447	340.5	317.1	
			317.0	
June 1	1420	316.0	316.4	0.4
		316.0		0.4
June 15	1353	316.5	316.4	−0.1
		316.5		−0.1
July 1	1402	316.5	316.6	0.1
		316.8		−0.2
July 15	1248	316.3	316.0	−0.3
		316.3		−0.3
Aug. 1	1515	313.6	312.8	†
		313.7		†
Aug. 15	1440	316.0	313.0	
		313.1		
Sept. 1	1130	313.5	314.1	0.6
		313.7		0.4
Sept. 15	1005	312.5	312.0	−0.5
		312.6		−0.6
Oct. 2	1200	312.5	312.7	0.2
		312.5		0.2
Nov. 3	1050	313.1	313.0	−0.1
		312.9		0.1
Nov. 15	1425	313.1	313.3	0.2
		313.1		0.2
Dec. 5	1405	315.1	315.4	0.3
		314.8		0.6
Dec. 15	1435	318.2	314.5	†
		318.2		†
1962				
Jan. 1	1147	314.5	314.4	−0.1
		314.8		−0.4
Jan. 15	1422	313.4	314.4	1.0
		313.7		0.7
Feb. 1	1215	315.9	315.1	−0.8
		316.6		−1.5
Feb. 15	1015	315.5	315.6	0.1
		315.5		0.1
March 1	1235	316.7	316.4	−0.3
		316.7		−0.3
March 15	1435	317.0	317.2	0.2
		317.4		−0.2
April 1	1520	318.6	316.9	
		317.1		
April 16	1525	317.4	316.7	−0.7
		317.5		−0.8
May 1	1050	318.4	318.0	−0.4
		318.4		−0.4
May 15	1405	318.8	318.6	−0.2
		318.8		−0.2
June 1	1835	318.0	318.1	0.1
		317.9		0.2
June 15	1420	316.7	316.9	0.2
		317.0		−0.1
July 1	1615	316.0	315.6	−0.4
		316.1		−0.5
July 16	1105	316.8	316.7	−0.1
		316.8		−0.1
Aug. 1	1130	315.1	315.3	0.2
		314.8		0.5
Aug. 15	1500	314.1	314.0	−0.1
		314.4		−0.4
Sept. 15	1150	312.7	312.7	0.0
		312.7		0.0
Oct. 3	1410	311.8	311.6	−0.2
		311.7		−0.1
Nov. 2	1100	313.3	312.7	−0.6
		313.3		−0.6
Nov. 19	0955	314.5	313.9	−0.6
		314.4		−0.5
Dec. 2	2155	314.8	314.1	−0.7
		314.8		−0.7
Dec. 16	2210	315.8	314.9	−0.9
		315.8		−0.9
Average	1960 (17 flasks)			−0.5
	1961 (28 flasks)			−0.0
	1962 (42 flasks)			−0.2
Grand average				−0.2

* Computed only if two or more flasks taken at a given time vary in analysis by less than 1.0 ppm.

† Omitted because flasks samples were taken downwind from observatory.

TABLE 3. Concentration of Atmospheric Carbon Dioxide near Hilo, Hawaii, in Samples Collected in Glass Flasks

Date	Local Time	No. of Flask Samples	Average Concentration, ppm	Range, ppm
1960				
March 30	1800	3	316.3	0.41
March 31	Missing	3	316.4	0.17
July 3	0900	2	316.4	0.00
July 15	1400	2	314.8	0.61
Aug. 4	1530	2	312.7	0.12
Aug. 18	1430	2	310.1	0.40
Oct. 6	1520	1	311.3*	2.36
Nov. 4	1250	2	312.0	0.17
Nov. 25	1300	2	312.9	0.09
Dec. 1	1050	2	315.0	0.33
Dec. 20	1545	2	315.0	0.55
1961				
Jan. 4	1315	2	314.7	0.00
Feb. 2	1345	2	315.7	0.11
Feb. 17	1115	2	315.1	0.00
March 1	1525	1	316.0*	25.83
March 15	1525	2	317.4	0.11
April 4	1110	1	317.9*	1.36
May 15	1445	2	317.9	0.00
June 1	1425	2	318.7	0.56
June 15	1350	2	317.9	0.00
July 1	1356	2	317.3	0.67
July 15	1255	2	316.3	0.61
Aug. 1	1520	2	313.9	0.79
Aug. 15	1442	2	314.0	0.89
Sept. 1	1130	2	311.4	0.78
Sept. 15	1020	1	309.4*	1.56
Oct. 2	Missing	2	313.0	0.78
Oct. 15	1055	1	313.9*	1.79
Nov. 3	1051	2	315.3	0.00
Nov. 15	1425	2	315.2	0.10
Dec. 19	1440	2	314.5	0.00
1962				
Feb. 1	1215	2	318.2	0.11
Feb. 17	1430	2	316.3	0.11
March 1	1230	2	314.9	0.56
April 16	1520	2	318.9	0.58
May 5	1125	2	320.2	0.00
May 15	1410	1	318.6	
June 15	1307	2	317.9	0.24
July 1	1042	2	315.9	0.23
July 18	1400	1	311.0*	7.23
Aug. 1	1130	1	313.2*	2.04
Aug. 15	1350	2	313.7	0.90
Sept. 18	1300	1	312.6*	9.39
Nov. 1	1045	2	313.7	0.90
Dec. 4	1430	2	315.8	0.00
Dec. 14	1225	2	315.5	0.36

* Higher sample rejected.

We can also assess the data by comparing the seasonal trend at Mauna Loa with that for the vicinity of Hilo and that obtained from aircraft (Table 6 and Figure 14). To make such a comparison we have combined measurements for different years. Noting that the long-term rise at Mauna Loa is very close to the rate of 0.06 ppm/month (See Figure 11), we adjusted the values of Table 1 to a datum of January 1960 and computed averages for each calendar month. Averages for the vicinity of Hilo were obtained from the values of Table 3, similarly adjusted. As was done in comparing the Mauna Loa flask samples with the analyzer values to minimize the influence of contamination, we used only values based on duplicate or triplicate samples which agreed within 1.0 ppm. In Figure 14 the monthly averages have been plotted versus the weighted average date of sampling to take into consideration the variability in sampling dates. The values from aircraft sampling derived from Table 5, after adjustment, were plotted directly versus the actual dates of sampling.

The average seasonal course of CO_2 at Mauna Loa is very smooth. The values for the vicinity of Hilo, based on only a few pairs of flask samples per month, scatter more and tend to be above values for Mauna Loa by about 1 ppm. The aircraft values are also slightly higher than those for Mauna Loa, but neither the values for the vicinity of Hilo nor the aircraft values are significantly higher.

When we consider long-term variations, all differences between sampling locations are insignificant. The steady record at Mauna Loa, which indicates an average rise of 0.68 ppm/yr (see Figure 11), must indeed reflect the rise of CO_2 concentration in the undisturbed atmosphere at all levels up to 700 mb near the Hawaiian Islands.

Volcanic and biological influences. Short-term variations in concentration of CO_2 produced by volcanic and biological activity on Hawaii are a distinct feature of the continuous record at Mauna Loa Observatory.

Volcanic CO_2 can come either from a rift zone southeast of the observatory or from vents in the caldera to the south-southwest. The rift zone, being closer to the observatory, might be expected to produce most of the observed contamination. The zone consists of a line

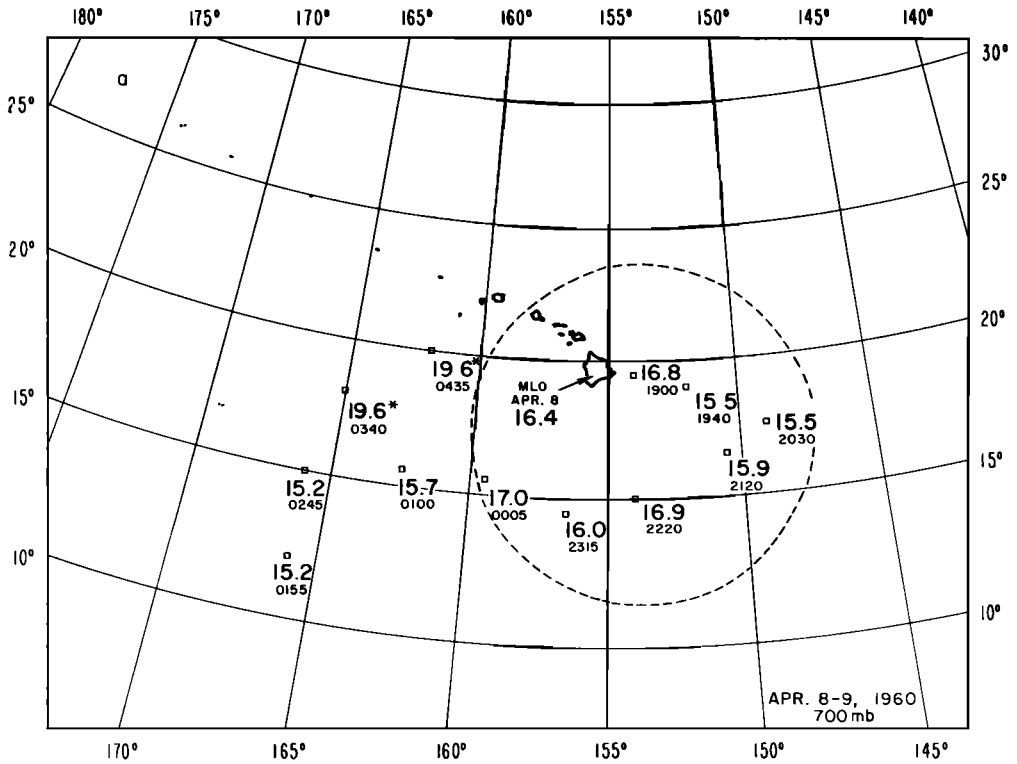


Fig. 12*a*.

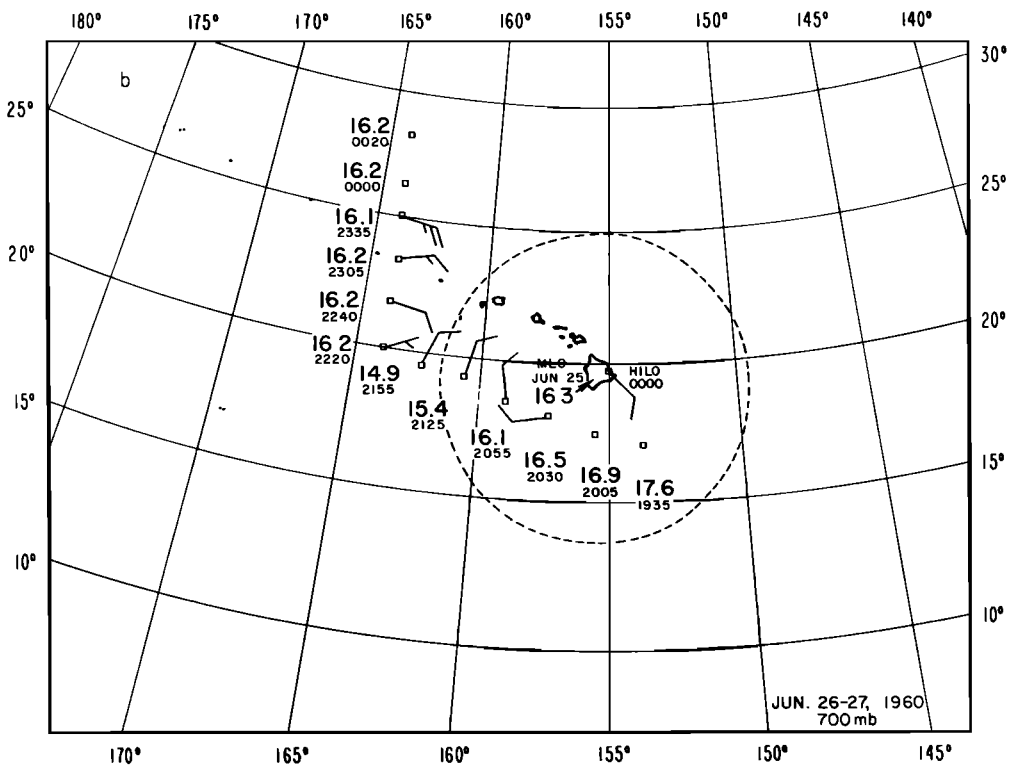


Fig. 12*b*.

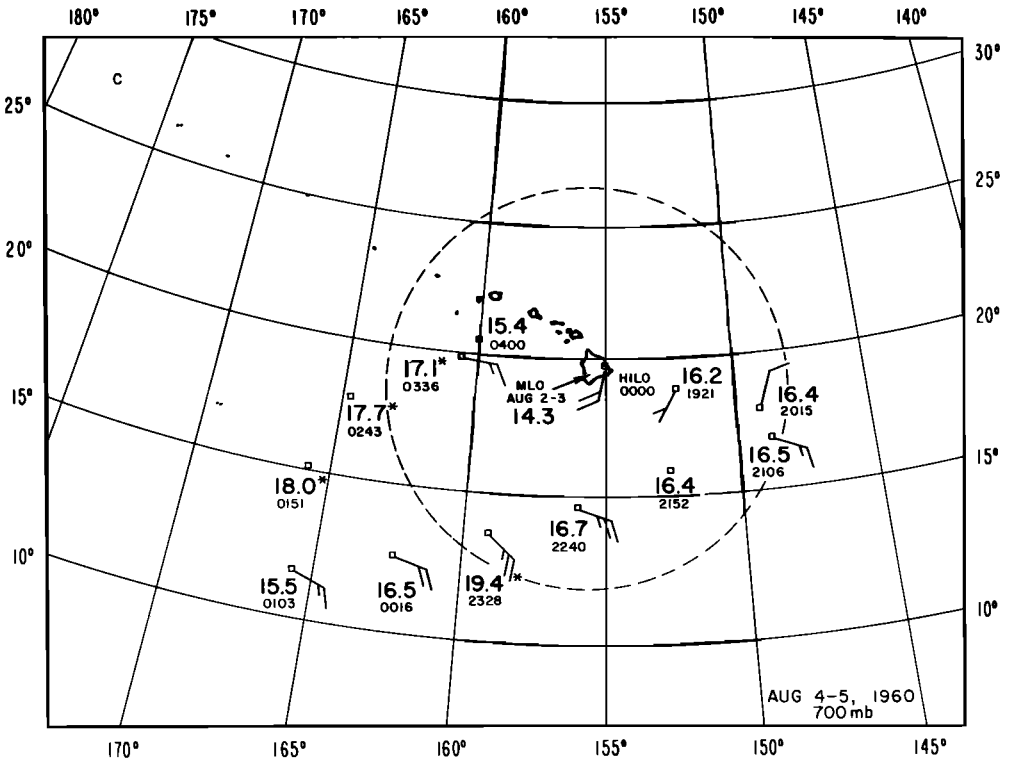


Fig. 12*c*.

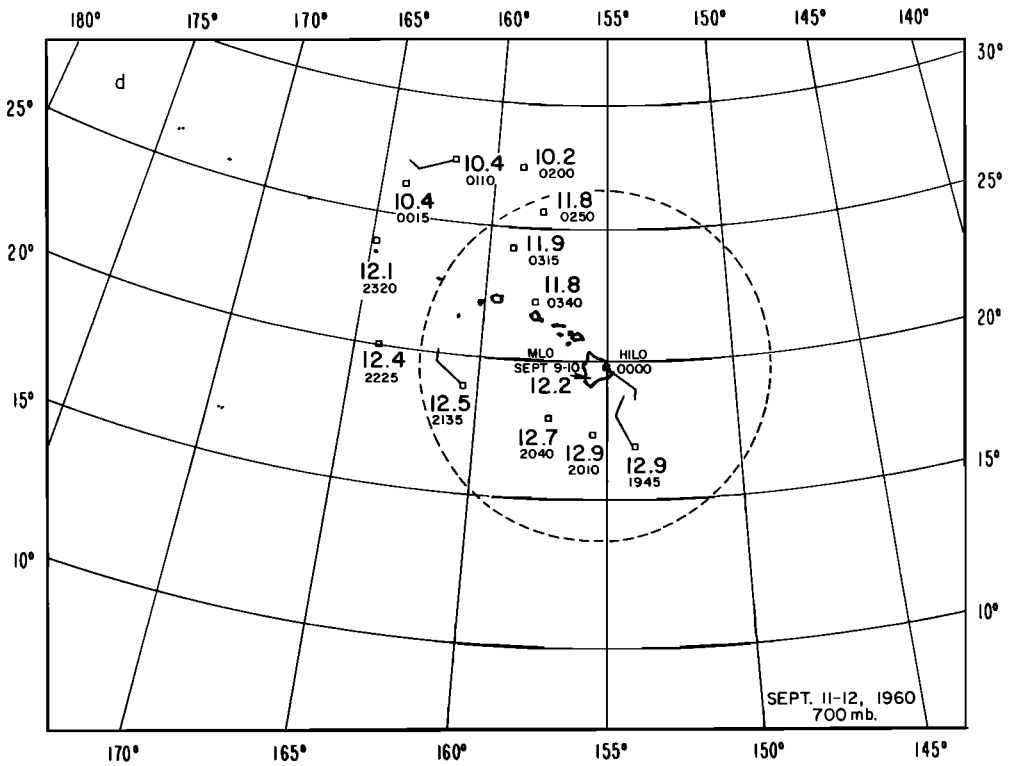


Fig. 12*d*.

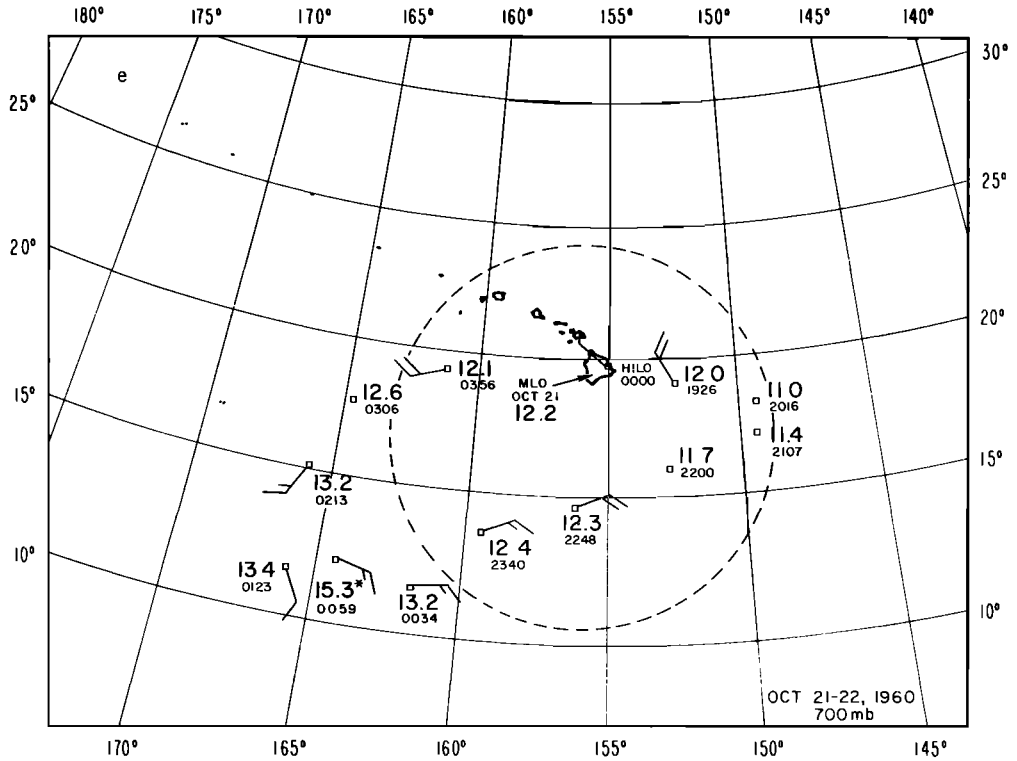


Fig. 12*e*.

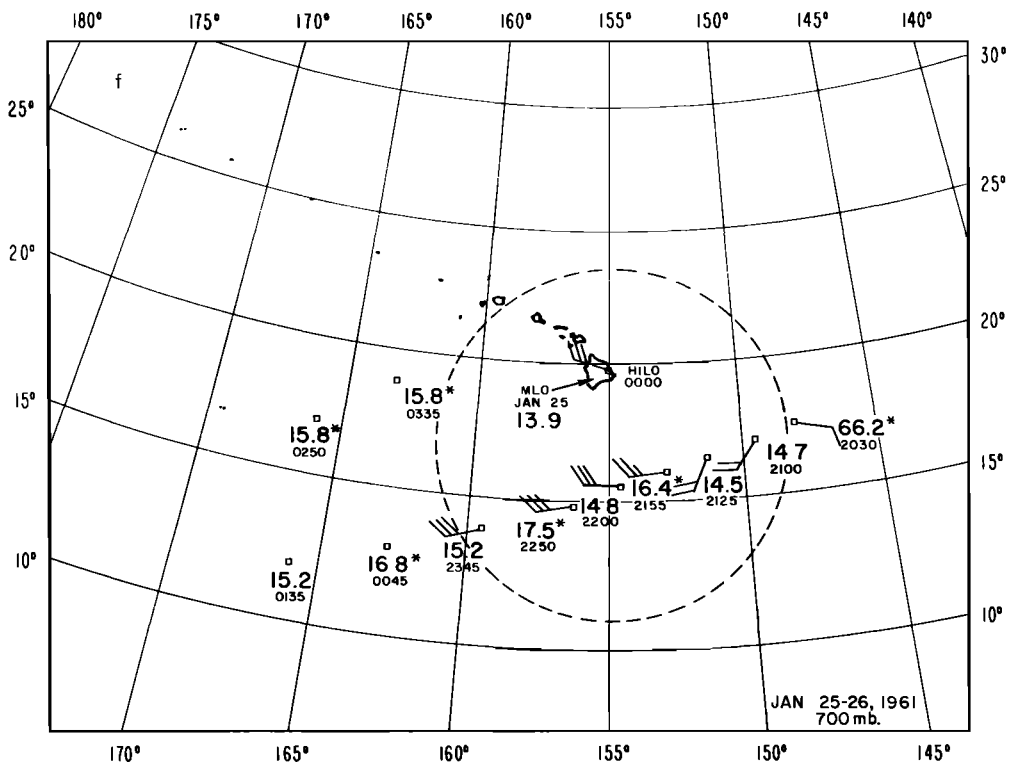


Fig. 12*f*.

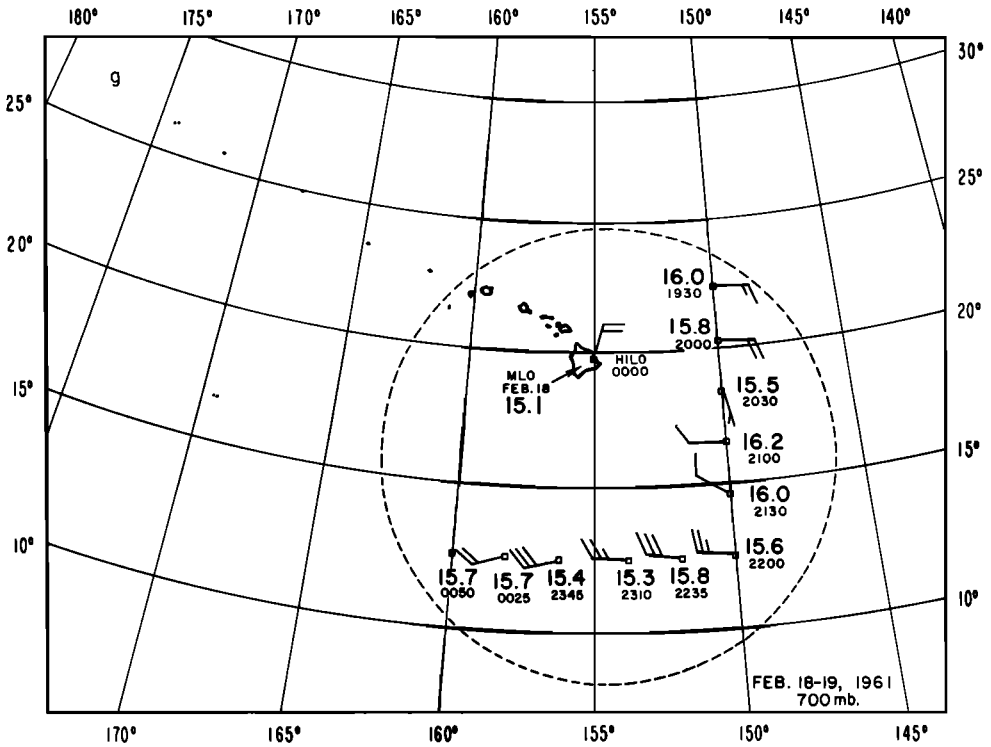


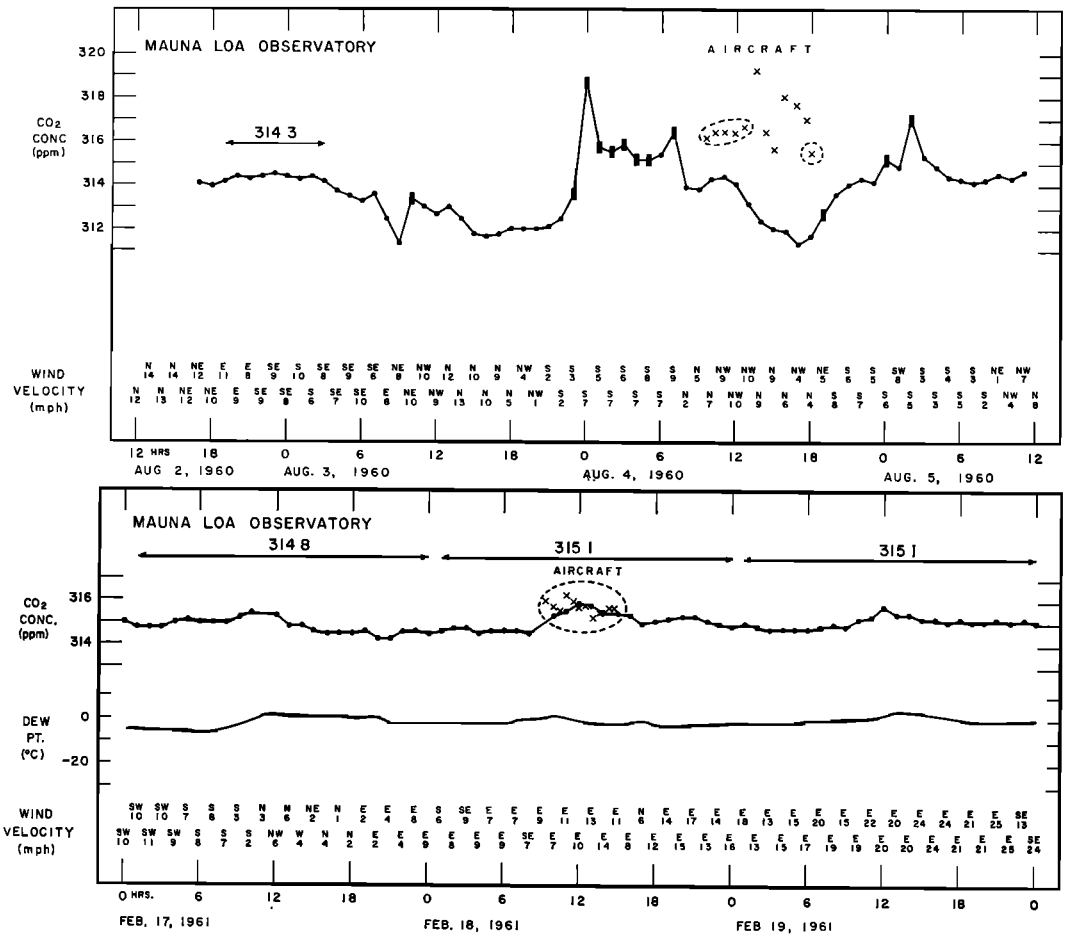
Fig. 12*g*.

Figs. 12*a* to 12*g*. Areal plot of the concentration of atmospheric CO_2 at the 700-mb pressure level on Loon Kilo flights of the U. S. Air Force. Positions are shown by squares. Wind velocity on the Beaufort scale is shown by arrows. Concentrations in parts per million appear with the first digit dropped, e.g. '15.5' is for 315.5 ppm. All values are based on single flask samples except those for Mauna Loa Observatory which are averages of the data from the gas analyzer for periods of steady concentration near the time of aircraft sampling. For the aircraft data time (UT) of sampling is shown below the concentration. For Mauna Loa Observatory the date of sampling is shown. Dashed circles enclose positions judged best to represent the atmosphere near Mauna Loa. Because the flight tracks varied the selection is somewhat arbitrary. Samples judged to be contaminated by cabin air are identified by asterisks and are not considered in the computations.

of vents about 10 km long about 4 km perpendicularly from the observatory. *McCormick* [1957] calculated the contamination expected to occur at the observatory by using an estimate of J. Eaton that the vents, when active, would release 1500 g/sec of CO_2. Assuming the effect of these at the observatory to be equivalent to an infinite line source with a mean emission rate per meter, Q, equal to 0.15 g/sec (the total emission divided by 10 km) at a distance normal to the source, x, of 4000 m, the ground level concentration of contamination, χ, was calculated for a wind direction normal to the line by the equation of *Sutton* [1953, p. 288]: where $\bar{u}$ is the average wind speed in meters

$$\chi = \frac{2Q}{(\pi)^{1/2}\bar{u}C_z x^{1-n/2}}$$

per second and C_z and n are special parameters depending on wind speed, height above ground, and the vertical temperature gradient. Choosing $\bar{u} = 10$, $C_z = 0.30$, $n = 0.25$, values appropriate to neutrally stable air in a moderately strong wind, McCormick found $\chi = 4 \times 10^{-5}$ g/m^3. This result, equivalent to an increase in atmospheric CO_2 concentration of 0.03 ppm, is over 100 times lower than observed, but if figures are substituted in Sutton's equation which are appropriate to a nocturnal mountain



Figs. 13*a* and 13*b*. The hourly average concentration of atmospheric CO_2 at Mauna Loa Observatory versus local time for selected periods during August 1960 and February 1961. Crosses denote contemporaneous aircraft samples. Dashed circles enclose values of aircraft samples whose positions, shown in Figures 12*a* to 12*g*, are judged best to represent the atmosphere near Mauna Loa. Other symbols are as in Figures 5*a* and 5*b*. Dew point data are not available for August 1960.

TABLE 4. Comparison of Data from Analyzer at Mauna Loa Observatory with Flask Samples Collected from Aircraft

Date		No. of Flask Samples	Average CO_2 Concentration, ppm		Difference
Aircraft	Mauna Loa		Aircraft	Mauna Loa	
	1960				
April 8	April 8	7	316.2	316.4	0.2
June 26	June 25	5	316.5	316.3	−0.2
Aug. 4	Aug. 2–3	6	316.3	314.3	−2.0*
Sept. 11	Sept. 9–10	7	312.4	312.2	−0.2
Oct. 21	Oct. 21	7	311.8	312.2	0.4
	1961				
Jan. 25	Jan. 25	4	314.8	313.9	−0.9
Feb. 18	Feb. 18	11	315.7	315.1	−0.6
Average (6 sampling periods)					−0.2

* Omitted from average.

TABLE 5. Analyzer Values at Mauna Loa Observatory for Selected Periods of the Day

Month	Monthly Average CO_2 Concentration, ppm		
	0–12 Hours	0–24 Hours	Steady Periods
1961			
April	316.64	316.40	316.36
May	317.60	317.39	317.37
June	316.82	316.69	316.62
July	315.40	315.03	315.70
Aug.	314.05	313.60	313.80
Sept.	312.54	312.17	312.14
Oct.	312.67	312.49	312.53
Nov.	313.24	313.14	313.09
Dec.	314.24	314.16	314.06
1962			
Jan.	315.28	315.08	314.97
Feb.	315.54	315.44	315.48
March	316.80	316.60	316.60
Average	315.07	314.85	314.89

TABLE 6. Monthly Average Concentration of Atmospheric Carbon Dioxide (ppm) Referred to a Constant Datum (January 1960)

Month	Near Hilo	Mauna Loa Observatory	At 700 Millibars near Hawaii
Jan.	314.0	313.5	314.1
Feb.	315.2	314.0	314.9
March	315.7	314.8	
April	317.3	315.8	316.0
May	317.7	316.4	
June	316.9	315.9	316.2
July	315.2	314.3	
Aug.	311.9	312.6	315.9
Sept.	310.2	311.0*	311.9
Oct.	311.7	310.7	311.3
Nov.	312.6	311.8	
Dec.	313.8	312.6	

* September 1958 value omitted from average because value is based on data for only 3 days.

wind during strong temperature inversion [*Gifford et al.*, 1955, p. 54], i.e., $C_s = 0.07$, $n = 0.50$, and $\bar{u} = 3$, then $\chi = 2 \times 10^{-8}$, equivalent to 2 ppm, which is reasonable. Without a detailed micrometeorological study, a more refined calculation is not justified. The result indicates that contamination at night is probably transported in a relatively shallow layer of air, such as *Geiger* [1959, pp. 241–247] described for a conical mountain.

The layer of air which produces the afternoon dips may be considerably thicker. According to *Becking* [1962] a fast-growing tropical forest can produce wood at the rate of 50 m^3/hectare yr. If we assume the density of wood to be 0.5 and the carbon content 50% by weight, the flux of CO_2 needed to sustain this rate of growth is 1.6×10^{-7} g/cm^2 sec. A stream of air passing over such a forest for 2 hours would lose 10^{-3} g/cm^2 in the vertical column, enough to deplete a layer of air 1 km thick by 10 ppm. Because the forests of Hawaii are not harvested, much of the CO_2 needed to sustain growth may be supplied through a local cycle involving oxidation of accumulated dead plant material within the forest itself. But even if only the order of 1% of the CO_2 for new growth comes from the upslope winds, this is sufficient to produce the dips observed at Mauna Loa Observatory through an air layer of the order of 100 m in thickness.

The annual average diurnal course of CO_2 (Figure 8) indicates that the average afternoon dip persists into the night, well beyond the time when the normal upslope wind is replaced by a downslope wind. If the above estimate of depletion by forests can be accepted, the updraft on Mauna Loa during the day must on average carry a very substantial amount of depleted air to the upper slopes of the mountain before the nocturnal regime sets in. But, since the night circulation probably involves a shallow layer of air, several hours, at least, may elapse before the depleted air near the summit can be replaced by undepleted air from aloft. The delay should be greatest when the general horizontal winds above the trade wind layer are weak and should nearly disappear when they become sufficiently strong.

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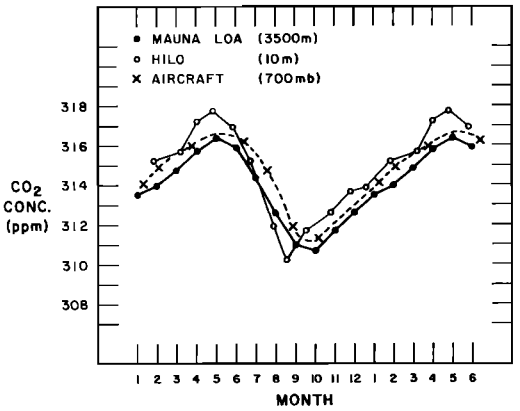

Fig. 14. The concentration of atmospheric CO_2 at Mauna Loa Observatory, near Hilo, and at 700 mb near Hawaii as a function of the month of the year. Values have been referred to a January 1960 datum. January through June (months 1 to 6) are plotted twice to reveal the seasonal pattern more fully.

version. Personnel of the 55th Weather Reconnaissance Squadron of the United States Air Force collected the aircraft samples under the guidance of Major Bernard M. Rose and Captain E. B. Huber. Messrs. Harry Arashiro, Howard Ellis, Colby Foss, Cliff Kutaka, and Robert Williams of the staff of Mauna Loa Observatory operated the gas analyzer, processed data, and collected flask samples at the observatory and near Hilo. Messrs. Eugene Collias, Frank Maurer, Lee Waterman, Mrs. Denise Phleger, and Mrs. Joan Soutar of the Scripps staff and Dr. Eugene Wilkins and Mr. Thomas Harris of the U. S. Weather Bureau further processed the data. Mr. Stig Fonselius assisted on many phases of the program at Scripps while on leave from the International Institute of Meteorology, Stockholm. Mr. Harris and Mrs. Blanche Christman analyzed the flask samples.

The inauguration of the program was aided by the Departments of Oceanography of the University of Washington and Texas Agricultural and Mechanical College, the Lamont Geological Observatory, and Woods Hole Oceanographic Institution, who generously donated equipment purchased from their own funds for the International Geophysical Year.

The Scripps program to monitor CO_2 in the atmosphere and oceans was conceived and initiated by Dr. Roger Revelle who was director of the Scripps Institution of Oceanography while the present work was in progress. Revelle foresaw the geochemical implications of the rise in atmospheric CO_2 resulting from fossil fuel combustion, and he sought means to ensure that this 'large scale geophysical experiment,' as he termed it, would be adequately documented as it occurred. During all stages of the present work Revelle was mentor, consultant, antagonist. He shared with us his broad knowledge of earth science and his appreciation for the oceans and atmosphere as they really exist, and he inspired us to keep in sight the objectives which he had originally persuaded us to accept.

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