Variations of the carbon dioxide content of the atmosphere in the northern hemisphere

By BERT BOLIN and WALTER BISCHOF, *Institute of Meteorology, University of Stockholm*

(Manuscript received March 4, 1970; revised version May 28, 1970)

ABSTRACT

Six years of measurements (1963–1968) of carbon dioxide in the troposphere and the lower stratosphere are presented. The data reveal an average annual increase of the CO_2-content of 0.7 ± 0.1 ppm/year, while during this time the annual industrial output has increased from about 1.9 ppm to 2.3 ppm/year. Thus the increase in the atmosphere is about $\frac{1}{3}$ of the total output. Considerations of the possible increase of vegetative assimilation due to the higher CO_2-content of the atmosphere reveals that this is at most $\frac{1}{4}$ of the output, probably considerably less. The net transfer to the oceans thus is at least equal to $\frac{1}{2}$ of the industrial output. The transfer rate across the sea surface seems effective enough not to represent an appreciable resistance and the decisive factor for determining this transfer therefore is the ocean circulation or turn over rate. The figures quoted indicate that 20–25 % of the world oceans must have been available during the time of rapid increase of the industrial output of CO_2 (the last 30–50 years) to explain the rather large amount that has been withdrawn from the atmosphere. Still a continued increase of the fossil fuel combustion as forecast by OECD implies that the CO_2-content of the atmosphere at the end of the century will be between 370 ppm and 395 ppm as compared with 320 ppm, the average value for 1968.

The amplitude of the seasonal variation is found to be about 6.5 ppm at 2 km and 3.5 ppm in the uppermost part of the troposphere. The phase shift of the seasonal variation between these two levels is 25–30 days. On the basis of these data a vertical eddy diffusivity $K = 2 \cdot 10^5$ cm^2 sec^{-1} is derived. The amplitude of the seasonal variation in the lower stratosphere, 11–12 km, is less than 1 ppm and the phase is delayed at least $1\frac{1}{2}$ month as compared with the upper troposphere.

1. Introduction

The combustion of fossil fuel is increasing at an accelerated rate and so is therefore the output of carbon dioxide into the atmosphere. In view of the importance of carbon dioxide for radiative processes in the atmosphere and thus for its heat balance, the continuing increase of CO_2 in the atmosphere has been the subject for many investigations. Insufficient accuracy of the measurements long made it difficult even to ascertain the magnitude of a probable increase. Also, the measurements were often not representative for the main part of the atmosphere due to local sources and sinks of considerable intensity (vegetation, soil release, combustion). Fonselius et al. (1956) attempted a survey of available observations since the middle of the last century but could only conclude that it seemed as if by far the greatest part of the CO_2 released by human activities, at that time 14–15 % of the total CO_2-content of the atmosphere, had remained in the atmosphere. Callendar (1958) in a similar study also concluded that the measurements in 1955 and 1956 lie "almost on the fuel line", 12 % above the 1900-base value of 290 ppm. On the other hand Revelle & Suess (1957) expressed the opinion "that most of the CO_2 released by artificial fuel combustion since the beginning of the industrial revolution must have been absorbed by the ocean", based on the C^{14}-variations in wood over the last 100 years. Bolin & Eriksson (1959) showed, however, that the C^{14}-content of wood was compatible with an assumption that between

---

This work has been supported by the Swedish Science Research Council as part of Contract No. 2065-16.

Table 1. *Distribution of observations according to latitude and height*

	South of 30° N	30°–50° N	50°–70° N	70°–90° N
1–3 km	—	—	124	3
3–7 km	5	1	87	4
7–9 km	—	2	70	12
above 9 km	30	33	269	61
Sum	35	36	550	80

¼ and ¾ of the CO_2 output remained in the atmosphere. Keeling (1960) and later Bolin & Keeling (1963) verified this result when they found that measurements in regions far from local sources and sinks showed an annual increase of CO_2 of 0.6–0.7 ppm as compared with 1.4 ppm to be expected at that time from the combustion of fossil fuel, if no removal from the atmosphere took place.

2. Sampling programme

In view of the importance of settling more definitely the magnitude of CO_2-increase, which might also permit more accurate forecasts of the likely increase during the remainder of this century, it was decided in 1962 to initiate a long term sampling programme and to make systematic attempts to improve the accuracy of the analysis. The method now used for sampling and the technique employed for analysis is described separately by one of us (Bischof, 1970). We merely recall here that the present relative accuracy is 0.2 ppm. The absolute accuracy is considerably less. The standard used is the same as the one to which Keeling (1960), Bolin & Keeling (1963) have referred their analyses and our data are therefore comparable. On the basis of absolute determinations by Keeling an annual average increase of the Scripps standard of about 0.06 ppm seems to have securred over the last ten years (personal communication). Such a correction has tentatively been applied to all data. For the more detailed intercomparisons between the Stockholm and Scripps standards reference is made to a later publication.

Since there exist large sources and sinks of carbon dioxide at the earth's surface, it was decided to sample primarily the upper troposphere and lower stratosphere in order to get as representative measurements for the bulk of the atmosphere as possible. Air samples have been collected from commercial aircrafts on route over Scandinavia, between Scandinavia and North America and between Scandinavia and Japan flying across the Arctic Sea. A more limited number of observations are also available from the Indian Ocean, the Caribbean Sea and the northern parts of South America, but these data will not be included in this presentation. All samples have been carefully checked for leakage or contamination and altogether 701 good samples from about 50 flights are included in the present analysis. Their distributions as a function of height and latitude and with regard to time is given in Tables 1 and 2. All data on which the present analysis is based are available on request in the form of a technical report.

It can be seen from these tables that the data are rather unevenly distributed and that coverage is not quite as adequate as would have been desirable. It has been possible, however, to intensify the sampling during 1969, the results of which will be reported later.

It soon became clear that the data showed

Table 2. *Distribution of observations in time*

	J	F	M	A	M	J	J	A	S	O	N	D	Year
1962	—	—	—	—	—	—	—	—	—	—	—	17	17
1963	—	9	—	10	53	5	18	60	39	92	10	9	305
1964	10	—	—	28	8	10	—	—	—	16	—	20	92
1965	—	—	—	23	—	—	—	—	—	—	25	—	48
1966	24	14	46	11	—	18	—	—	—	—	—	—	113
1967	—	—	—	7	—	38	—	—	—	13	—	—	58
1968	39	—	—	—	—	—	—	—	29	—	—	—	68
Sum	73	23	46	79	61	71	18	60	68	121	35	46	701

many interesting features in addition to the annual increase. Some preliminary results have been published before (Bischof, 1965; Bischof & Bolin, 1966). In addition to a possible annual increase of the CO_2-content of the atmosphere, we shall here also discuss the seasonal variations and the variability and their height dependence.

3. Data analysis

We may consider the observed variations of the carbon dioxide content of the atmosphere as a result of

(*a*) an annual increase

(*b*) seasonal variations which are height and latitude dependent (cf. Bolin & Keeling, 1963)

(*c*) irregular variations, which also vary with latitude and elevation, due to the mixing processes constantly at work in the free atmosphere and sometimes clearly associated with fronts or other characteristic features of weather systems.

The present data hardly permit a search for possible secular variations in the annual increase. It was therefore assumed that the rate of increase remained constant over the six years covered by the present analysis. Some further comments on this question will be given below. The seasonal variations were also assumed to be the same from one year to the next, although it is quite conceivable that some differences may exist. Since about 80% of the data were collected between 50° N and 70° N, the latitude dependence of the annual cycle of CO_2 variations could not be studied.

Table 3. *Seasonal CO_2 variations (deviations from annual mean) at different elevations in the troposphere (ppm)*

	Elevation, km				
Month	1–3	3–5	5–7	7–9	9–tropopause
J	2.0	1.6	1.3	0.7	0.6
F	3.5	2.3	2.0	1.6	1.3
M	5.2	3.2	2.7	2.3	2.0
A	6.5	3.8	3.4	3.0	2.7
M	2.5	3.6	3.8	3.7	3.4
J	−0.7	0.4	1.0	1.7	3.1
J	−4.0	−3.2	−2.4	−1.9	−1.4
A	−7.0	−5.7	−5.0	−4.3	−3.6
S	−8.5	−6.7	−4.7	−3.9	−3.6
O	−1.5	−1.4	−2.2	−2.6	−2.4
N	0.2	0.3	−0.5	−1.4	−1.4
D	1.3	1.4	0.8	0.4	0.1
Range of variations	15.0	10.5	8.8	8.0	7.0

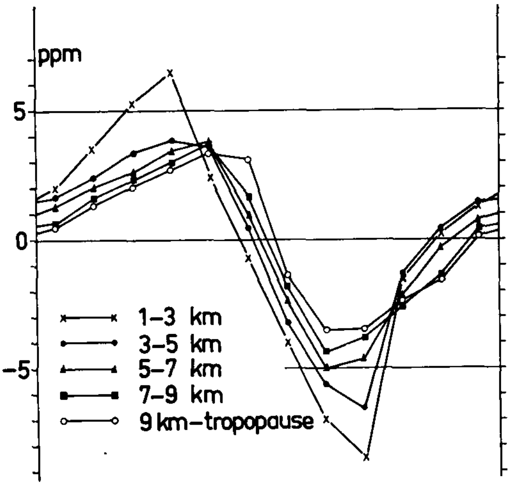

Fig. 1. Seasonal variations of the CO_2 content of the atmosphere at different levels in the troposphere.

Preliminary visual inspection of the data showed clearly that the seasonal variations decrease with elevation and very much so when proceeding into the stratosphere. Therefore, first the data from the *troposphere* were grouped according to height in the intervals 1–3 km, 3–5 km, 5–7 km and 9 km–tropopause. Occasionally it is of course difficult to tell whether a sample is from the troposphere or not and in such cases the data were not included in the tropospheric analysis. All data were reduced to 1963 values, by assuming that the annual CO_2 increase was 0.5, 0.6, 0.7 and 0.8 ppm. Next the annual cycle was computed in these four cases, as well as the standard deviation of the individual values from the normalized monthly means. A minimum standard deviation was obtained when the annual increase was assumed to be 0.7 ppm and a closer analysis yielded a value of 0.72 ± 0.1 ppm as the most likely average value for the annual increase during the period considered, i.e. December 1962 to September 1968. If all data are reduced to 1 January 1963 with due regard taken to such an annual increase and if the mean seasonal variations are eliminated we obtain an average value for the CO_2-content of the atmosphere

Table 4. *Amplitude (A) and phase (α) of annual and semi-annual oscillation of the CO_2-content of the atmosphere*

	Annual		Semi-annual	
Height, km	A_1 (ppm)	α_1(°)	A_2 (ppm)	α_2(°)
1–3	5.7	26°	2.0	192°
3–5	4.3	25°	1.8	188°
5–7	3.7	15°	1.6	174°
7–9	3.4	5°	1.2	174°
9–tropopause	3.2	− 2°	1.2	168°

on that date of 316.0 ppm. If we accept the mean value for the latitude belt north of 50° N for 1 January 1960 as given by Bolin & Keeling (1963) to be 314.0 ppm, we find an annual increase for the three years 1960 to 1962 of 0.7 ppm. This value for the annual increase and the one obtained for the six years 1963 to 1968 agree well with the value established by Keeling (1960) using measurements in the Antarctica during 1958 and 1959. These data thus do not justify the conclusion that there has been a significant change in the rate of the annual increase. Observations during several more years are necessary to permit a firm conclusion regarding this very important question.

The average seasonal CO_2 variations in the troposphere as deduced from the present data set are given in Table 3 and also shown in Fig. 1. It is seen that the range of the seasonal variations decreases markedly with elevation and that the phase is somewhat delayed at higher levels. Thus the maximum is reached during the first part of April at low levels, while it occurs towards the end of May near the tropopause. The minimum in late summer, on the other hand, seems to occur at approximately the same time, i.e. at the end of August or in early September. Table 2 shows however, that the data distribution is not very satisfactory to conclude definitely that this latter statement is true. A harmonic analysis of the seasonal variations is of some interest. Table 4 gives the amplitude and phase of the annual and semi-annual oscillation. The decrease of amplitude and change of phase as already obvious from Table 3 is here brought out explicitly.

Table 5. *Seasonal variations at different elevations as described by the first two harmonics given in Table 4*

	Elevation, km				
Month	1–3	3–5	5–7	7–9	9–tropopause
J	1.9	1.6	1.2	0.9	0.6
F	3.0	2.4	1.9	1.6	1.0
M	4.7	4.0	3.2	2.8	2.0
A	5.5	4.8	4.2	3.6	3.0
M	3.3	3.3	3.6	3.3	3.2
J	− 1.0	0.0	0.9	1.2	1.8
J	− 5.5	− 4.0	− 2.6	− 1.5	− 0.8
A	− 7.0	− 6.0	− 4.9	− 3.8	− 3.0
S	− 5.3	− 5.2	− 4.8	− 4.2	− 3.8
O	− 1.9	− 2.4	− 2.8	− 3.0	− 2.8
N	0.7	0.3	− 0.6	− 1.1	− 1.2
D	1.6	1.2	0.7	0.2	0.0
Range of variations	12.5	10.8	9.1	7.8	7.0

A marked change of the seasonal variation occurs when approaching the tropopause and proceeding into the stratosphere as was already indicated by Bischof & Bolin (1966). We consider first the data from levels well into the stratosphere and find that the CO_2-content of the atmosphere remains almost constant. Thus the first harmonic, obtained from all data above 11 km, the average elevation being 11.5 km and the maximum elevation 12.5 km, has an amplitude of only 0.6 ppm and a phase lag of about 80° behind the corresponding harmonic for the top layer in the troposphere (a maximum in August). This result is, however, hardly statistically significant since the scatter of the individual observations is relatively large.

In another analysis, in which a systematic attempt was made only to consider observations from the stratosphere, we also obtained a a slight annual oscillation even further delayed with a maximum in September. Again the results are not conclusive and we may at this stage therefore only conclude that there may be a slight seasonal oscillation of the CO_2-content of the air in the lower stratosphere, the amplitude of which, however, almost certainly is less than 1 ppm.

It is difficult to obtain an accurate mean value for the CO_2-content of the stratosphere for the reference time of 1 January 1963, because of the subjectivity that still is involved in classifying an air sample as "stratospheric" or "tropospheric". It seems clear that the mean value is lower than that for the upper

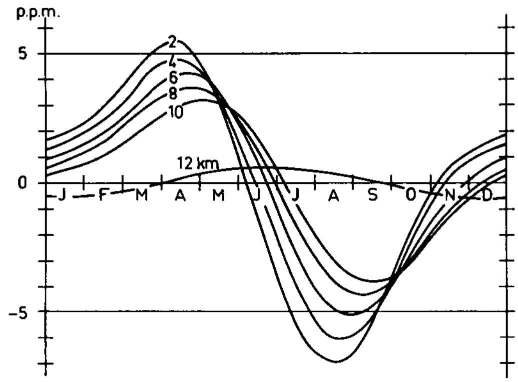

Fig. 2. Representation of the seasonal variations of CO_2 in the atmosphere at different levels with the aid of the first two harmonics.

troposphere, possibly by as much as 0.6 ppm. More observations from the stratosphere at all times of the year (summer values are very sparse in the present set) are required to permit a firm conclusion to be drawn and a supplementory programme is being initiated to achieve this.

Since the data hardly permit a significant analysis of more than two harmonics, it is of some interest to present the seasonal variations as obtained from only these two first harmonics. The result (Table 5 and Fig. 2) shows that a very regular change with height of amplitude and phase. The curves shown in this figure give the most representative picture of the seasonal oscillation that the present data material can reveal. It is interesting to note that the root mean square difference between the monthly mean values as given in Table 3 and the ones in Table 5 derived from the first two harmonics (and depicted in Fig. 2) on the average is 0.7 ppm as compared with an average root mean square variation of the monthly values around the yearly mean of about 3.2 ppm. The distribution with height of these quantities is given in Table 6, columns 5 and 3. 75–80% of these variations is thus caught with the first two harmonics of the seasonal variations.

Individual observations during the various flights of course scatter around the mean for the time of the year as deduced from the annual increase and seasonal variations as discussed above. Again these variations are comparatively large in the lower troposphere and decreases upwards (see Table 6, column 4 and Fig. 3). Due to the marked difference in the seasonal variations in the upper troposphere and lower stratosphere and the fact that the height of tropopause varies in association with synoptic disturbances the variations in the vicinity of the tropopause are somewhat larger than in the upper troposphere. They then again decrease in the stratosphere.

Since the flights in the upper troposphere and lower stratosphere often proceed at a constant elevation over long distances we have an opportunity to study how the spatial variations horizontally are dependent on the scale. We have computed the root-mean square of the difference in CO_2-content at two points as a function of the distance between them. The result as obtained for flights in the upper tropo-

Table 6. *Root mean square variations of the CO_2-content of the atmosphere (cf. text)*

(1) Height (km)	(2) Total variations of individual observations (ppm)	(3) Average seasonal variations (ppm)	(4) Variations of individual observations around monthly mean (ppm)	(5) Variations of monthly mean (ppm)
1–3	4.9	4.4	2.1	1.1
3–5	3.8	3.4	1.5	0.7
5–7	3.1	2.9	0.9	0.4
7–9	2.8	2.6	0.9	0.4
9-troposphere	2.6	2.4	1.0	0.5
Average for troposphere	3.5	3.2	1.4	0.7
Stratosphere (average height 11 km)	1.3	0.7	0.8	0.6

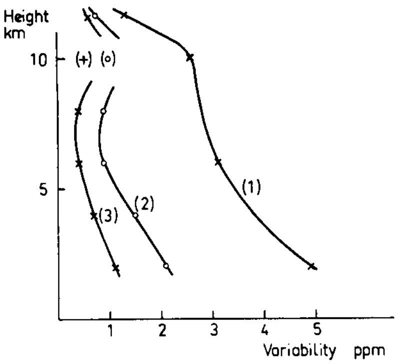

Fig. 3. Root mean square variations of the CO_2 content of the atmosphere as a function of elevation. *1*, Total variations of individual observations around the annual mean; *2*, variations of individual observations around the monthly mean; *3*, variations of the monthly mean.

sphere (above 9 km) is shown in Fig. 4. Even though the data amount is limited, a clear increase from a value of 0.6–1.0 ppm over distances up to about 750 km to about 1.5 ppm over 2 000–3 000 km is obtained. On the basis of a value of 1.0 ppm for the root mean square variation of the individual observations around the monthly mean in the upper troposphere (Table 6 column 4), we would expect an average value of this difference of 1.4 ppm, which is in good agreement with the result displayed in Fig. 4. It is clear that a considerable part of the total variation is found on comparatively small scales, but also that a not insignificant part is associated with the synoptic and large scale atmospheric systems. At least qualitatively this is similar to what is the case for atmospheric moisture.

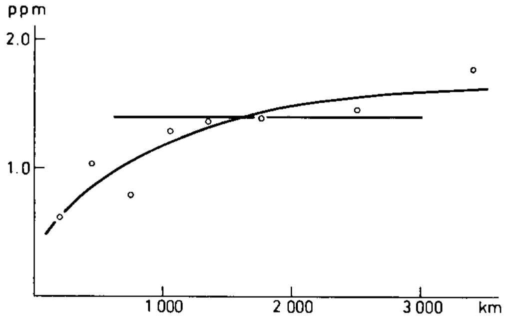

Fig. 4. Root-mean square of the difference of the CO_2 content of the atmosphere at two points as a function of distance between them (circles) at levels between 9 km and the tropopause.

Table 6 and Fig. 3 also give the total root mean square variations. It is clear that the seasonal variations are by far the most important factor in determining the variations of the CO_2-content of the atmosphere.

4. Discussion

4.1. THE ANNUAL INCREASE

4.1.1. *An assessment of present measurements*

As already mentioned in the introduction it has previously been estimated that an annual increase in atmospheric CO_2 of 0.6–0.7 ppm takes place. The present result agrees well with this earlier value. It should be noted, however, that the CO_2 production due to fossil fuel combustion in the meantime has increased considerably. The estimates by the United Nations, as summarized by Revelle & Suess (1957), are reproduced in Table 7. These values have recently been considerably revised by OECD (1966), see Table 7[1]. If all the CO_2 released to the atmosphere by mans activity remained in the atmosphere, the UN figures in Table 7 would correspond to an annual increase of 1.4 ppm and 1.7 ppm for 1960 and the period 1963–1968, respectively while the OECD estimates would imply an annual increase of 1.7 ppm and 2.1 ppm. The forecasts for later years differ even more. The main reason for this

Table 7. *CO_2 added to the atmosphere by fossil fuel combustion expressed in per cent of preindustrial amount in the atmosphere (~ 290 ppm) according to estimates by UN (1955) and OECD (1966)*

	Average amount added per decade, %			Cumulative total added at the end of decade, %		
	UN	OECD		UN	OECD	
1950–59	3.9	4.4		15.6	16.1	
1960–69	5.4	6.8		21.0	23	
1970–79	7.5	10.0		28.5	33	
1980–89[a]	10.5	15	16	39.0	48	49
1990–99[a]	14.5	22	26	53.5	70	75
2000–09[a]	20.0	33	42	73.5	103	117

[a] From 1980 the two values in the OECD-column refers to an annual increase of the CO_2-output by 4 % and 5 % respectively.

difference seems to be an assumption in the UN report, that the efficiency in the methods of utilizing the energy released by burning fossil fuel would increase considerably, whereby a less rapid increase of the use of fossil fuel would be required. This hardly seems to have been the case. It may be reasonable to assume that the values by OECD are more reliable, since data from about 10 more years were available. Still there is of course considerable uncertainty in any of the values from 1970 and onwards. The more detailed OECD forecast does not extend beyond 1980. Therefore, from 1980 two sets of values are given, one based on an annual increase of the CO_2 output of 4%, which is the mean value given by OECD for the decade 1970–1979, while the other set uses the value 5%, which is the mean for the two decades 1960–79. In both cases the expected increase is quite spectacular, particularly when considering that nuclear energy has been assumed to play an increasing role as an energy source when estimating the consumption of oil and coal.

In view of the fact that the CO_2-output over the last decade has increased by more than 50% as compared with the 1950's (6.8% and 4.4% respectively) while the annual increase in the atmosphere has remained almost the same, a considerably smaller percentage now seems to stay in the atmosphere than was the case in the late 1950's. Such a conclusion is of course critically dependent on the standard gas for calibration not having changed, as is the whole discussion of the CO_2-trend. A change of more than 1 ppm probably as much as 1.5 ppm, of this standard would be required in order to invalidate our conclusion. This is unlikely. However, previous estimates of the annual increase are quite uncertain and may well have been in error to the extent that we still are not justified in saying that the percentage staying in the atmosphere is decreasing.

The best possible conclusion that can be drawn from the present data therefore is that *35 ± 7% of the output of CO_2 due to fossil fuel combustion seems to stay in the atmosphere and that possibly a decrease of this percentage from a value above 40% may have occurred during the 1960's.*

Table 8. *Annual mean value of the CO_2 of the atmosphere 1960–1968 (referred to 1 July)*

Year	CO_2-content (ppm)
1960	314.3
1961	315.0
1962	315.7
1963	316.4
1964	317.1
1965	317.8
1966	318.5
1967	319.2
1968	320.0

If for a moment we assume that this percentage 35 ± 7% has remained constant since the burning of coal and oil began on a large scale, we extrapolate that the CO_2-content of the atmosphere in the middle of the last century should have been 296 ± 4 ppm. If it has decreased in recent years, as the observations possibly indicate the nineteenth century value should be correspondingly lower, yielding better agreement with earlier estimates of a preindustrial concentration of about 290 ppm. Again these earlier estimate are, however, quite uncertain. We conclude: *Of an accumulated input to the atmosphere until 1 January 1969 of about 22% of the preindustrial CO_2-content of the atmosphere, at least about 8% seems to have remained in the atmosphere.*

The best present estimates of the annual mean value of the CO_2-content of the Northern Hemisphere troposphere as obtained from measurements over the last decade are given in Table 8. All data have been referred to the standard gas being used by Keeling (Keeling, 1960; Bolin & Keeling, 1963).

4.1.2. *The role of the biosphere on land*

It is of interest to analyze the previous result somewhat closer. It has been customary to study the transfer of CO_2 between the atmosphere, the ocean and the biosphere with simple models (see i.e. Craig, 1957; Bolin & Eriksson, 1959). The exchange of Carbon-14 has been dealt with in a similar way (see also for instance Nydal, 1968; Young & Fairhall, 1968; Rafter & O'Brien, 1970). The atmospheric reservoir is

---

[1] *Note added in proof:* The figures for the 1960's are about 10% higher than those given in "Restoring the Quality of Our Environment. (US Environmental Pollution Panel, 1965) and additional data for 1963–1967 provided by Lester Machta, ESSA.

in exchange with the biosphere and the oceans. The size of the biosphere has been estimated as 50–85 % of the size of the atmosphere (Hutchinson, 1954; Eriksson & Welander, 1956; cf. also Rodin & Basilievič, 1968) and the annual assimilation to about 3 % of CO_2-content of the atmosphere. This corresponds to a residence time of CO_2 in the atmosphere before assimilation of 30–35 years. The major part of this assimilation is due to the forests (about ⅔). Gradually the CO_2 thus withdrawn from the atmosphere returns by decay of the dead organic material, humus and also by burning of the products from the lumber and pulp industries. It is well known that the rate of assimilation depends on the CO_2-amount in the atmosphere and one may therefore wonder how much of the CO_2 brought into the atmosphere by human activity has been absorbed by the vegetation and thus temporarily stored, essentially in the forests and in the humus.

If we assume that the CO_2 released to the atmosphere since 1880 can be described by $\gamma = A \exp [r(t - t_0)]$ (see Bolin & Eriksson, 1959), the accumulated amount at any time during this period is given by $G = A \cdot r^{-1}\{\exp [r(t - t_0)] - 1\}$. Let us for simplicity assume that the net increase in the atmosphere is given as a fraction, $\alpha(\approx 35\%)$, of this amount. We can obtain a maximum value for that part β of the CO_2 input to the atmosphere, which has been absorbed by the biosphere, if we assume that the assimilation increases directly proportionally to the amount of CO_2 in the atmosphere (Lundegårdh, 1957). In reality it probably is smaller, because the relation between CO_2 content of the atmosphere and growth is not linear and also because other factors such as temperature or moisture conditions may be limiting the growth rather than the CO_2-content of the atmosphere. Also, part of CO_2 that has been assimilated since 1880 is by now returning to the atmosphere through the decomposition of dead organic compounds. A computation of the accumulated absorption in the biosphere on the basis of the assumptions just mentioned and a residence time of CO_2 in the atmosphere before assimilation of about 35 years shows that a *considerable part, possibly as much as 15–20 % of the CO_2 released to the atmosphere by fossil fuel combustion, may have found its way into the biosphere*. It would be interesting to see if this could be verified by tree ring studies. An increase of 7 % between 1880 and 1968 should have taken place, or correspondingly less if other factors are limiting the growth. It should be emphasized that the increased assimilation is due to the accumulated excess CO_2 and that the return to the atmosphere of the assimilated CO_2 goes via the decomposition of dead organic material, a comparatively slow process. Therefore probably a somewhat larger *percentage* of the fossil CO_2 now is found in the biosphere than was the case a few decades ago. This circumstance may be one reason for a possibly slower increase of the CO_2 in the atmosphere in recent years, but data are lacking to permit a firm conclusion.

4.1.3. *Exchange with the mixed layer of the ocean*

Even though a significant increase of the biomass thus probably has taken place due to the fossil fuel combustion, about half of the industrial CO_2 released to the atmosphere seems to have gone into the oceans. It was pointed out by Bolin & Eriksson (1959) that it is necessary to consider the buffering effect of the CO_2 system of the sea in trying to study the transfer of CO_2 from the atmosphere to the sea. An increase of the CO_2-content of the sea will shift the chemical equilibrium and increase the partial pressure much more than the total content of carbon in the sea, provided the alkalinity remains unchanged. The latter we may assume to be valid over the relatively short time period with which we are concerned in comparison with the characteristic turn-over time of the oceans.

The oceans contain about sixty times as much CO_2 as does the atmosphere. We may express the buffering effect of the CO_2 system of the sea in a simplified manner by stating that the capacity of the sea to absorb CO_2 is only 1/10 of what one might expect by merely considering the size of the ocean reservoir. This circumstance is the main reason why we observe an appreciable increase of CO_2 in the atmosphere at all since otherwise the oceans would be able to absorb an amount well above 90 % of the CO_2 so far released to the atmosphere by fossil fuel combustion in spite of their rather slow turn-over rate.

In trying to estimate the transfer of CO_2 into the sea it has been customary to divide the sea into two reservoirs, one being the so-called mixed layer extending to about 75 meters depth, the other being the deep ocean (Craig, 1957). The former contains about as much carbon as the atmosphere but an equilibrium with the atmosphere is established by letting merely 1/10 of the fossil CO_2 be absorbed by it (see above). The exchange rate between the atmosphere and the mixed layer is characterized by an atmospheric residence time of about five years. This value is reasonably well established, particularly through studies of the decrease of bomb produced carbon-14 in the atmosphere since the large scale atmospheric tests in 1962 and 1963 (Nydal, 1968; Young & Fairhall, 1968). This residence time is rather short compared with the characteristic rate of increase of fossil fuel combustion. The mixed layer of the ocean therefore on the average is rather well in balance with the atmosphere. The small effective capacity of the mixed layer is also apparent from the result obtained by Bolin & Eriksson (1959), that the percentage of the fossil fuel CO_2 remaining in the atmosphere is almost independant of exchange rate between the atmosphere and the sea. It is rather the turn-over rate of the oceans as a whole that is important.

It is well-known that the solubility of carbon dioxide in sea water is temperature dependant (Eriksson, 1963). If changes of the temperature of the ocean surface layers take place over periods of years or decades, the time is too short for the $CaCO_3$ system to change appeciably. In such a case a 1°C increase of the temperature would increase the CO_2 partial pressure of the gas phase by 5–6%. A decrease of the average sea surface temperature of the world ocean by 0.1–0.2°C would therefore account for the apparent decrease of the portion of the industrial CO_2 release that stays in the atmosphere. Present information of the surface temperature of the oceans are inadequate to assess whether such changes occur or not over time periods of a decade. The figures quoted show, however, how sensitive the atmosphere—ocean CO_2 system is to rather small changes.

4.1.4. *The role of the deep sea*

The computations by Bolin & Eriksson (l.c.) also show that a transfer into the oceans of as much as 50% of the amount brought into the atmosphere by fossil fuel combustion cannot be accounted for with a simple two-box model of the oceans unless the residence time of water in the deep sea reservoir is only about 200 years. Measurements of carbon-14 in the deep sea show that the characteristic age of deep water in the Atlantic Ocean is about 600 years and in the Pacific and Indian Oceans more than 1 000 years. It is thus clear that the two-box model of the sea is a gross oversimplification of reality. It should be noted at this point that a similar result has been obtained by Rafter & O'Brien (1970). They find that the changes of the carbon-14 activity in the surface layers of the South Pacific Ocean also are incompatible with a simple two-box model having a slow exchange between the surface layers and a deep-sea reservoir. The reservoir "deep ocean" is by no means in reality a homogeneous body of water. It includes such different water masses as the "*bottom water*" slowly being formed in the Antarctic regions, the "*common water*" of the Pacific and Indian Oceans, the "*deep water*" and the "*intermediate water*" of the Atlantic Ocean. Some of these different water masses or parts of them are in more rapid exchange with the surface waters than is indicated by an *average* residence time for deep ocean water of about 1 000 years. In view of what has been said above, it would therefore be interesting to have a statistical representation of the water masses of the world oceans in terms of a distribution with regard to carbon-14 age i.e. similar to what has been done with regard to temperature and salinity by Cochrane (1958), Pollak (1958), and Montgomery (1958). The number of measurements is, however, insufficient for such a representation. A further subdivision of the deep sea into several reservoirs therefore becomes quite arbitrary.

The fact that release of CO_2 by fossil fuel combustion seems to have led to a larger increase of the CO_2 in the sea than in the atmosphere (about one half as compared with one third of the total output) permits, however, some qualitative conclusions. Since the effective capacity of the oceans to absorb CO_2 only is about six times that of the atmosphere (1/10 of 60, cf. above), *20–25% of the oceans must have been accessible in a time less than or about equal to the time over which the essential part of the industrial CO_2 output has taken place i.e. the last 30–50 years.* In such a case the remainder of the world oceans could have a slow rate of communication with the surface waters and thus with the atmosphere. Its behaviour

Table 9. *The likely values for the CO_2 content of the atmosphere (ppm) based on assumptions that 35% and 45% stays in the atmosphere and the fossil fuel combustion given in Table 7*

	35 % (ppm)		45 % (ppm)	
1970	321		321	
1980	332		335	
1990	348	349	355	356
2000	371	378	388	395
2010	403	418	430	450

would be almost irrelevant for the CO_2 changes that have occurred since the latter part of the 19th century.

In the light of the conclusion reached above it would be interesting to study the transfer of water masses in the oceans as we know them from standard measurements of temperature, salinity oxygen, etc. to see which parts of the world oceans could possibly play the role of absorbing the excess CO_2 in the way that seems necessary.

It is also intuitively clear that a more detailed study of the long term increase of CO_2 in the atmosphere, i.e. an extension of what has been done over the last 10 years and thereby a possible detection of variations in the amounts being transferred into the oceans, would yield information on the ocean circulations, but again only for those parts of the oceans that are contributing rapidly to the atmospheric CO_2 sink. It is worth pointing out in this context that the exchange of surface waters with the deep ocean water bodies is a very sluggish process and indeed very dependent on the particular ocean surface temperatures and salinities that develop in the winter. Thus variations in the intensity of the convection to deep layers in the Atlantic south of Greenland are known to occur. If major differences exist from one year to the next or over, say, a decade, they should be detectable as changes in the annual CO_2 increase in the atmosphere.

4.1.5. *The likely future increase*

The discussion above shows that we still know very little of how rapidly the industrial CO_2 disappears into the biosphere and the oceans. It is not unlikely that the annual CO_2 increase in the atmosphere may vary in time as also the observations during the last decade seem to indicate. Any more accurate prediction of the future increase therefore becomes quite uncertain and it should be emphasized that we merely can extrapolate on the basis of the more or less exponential increase of the industrial output of CO_2 to the atmosphere. If significant climatic changes would become apparent as a result of such an increase of atmospheric CO_2, it might of course turn out to be necessary to switch to other energy sources than fossil fuels. The immense investment involved makes it unlikely that any significant effects could be achieved before the last decade of this century.

Accepting 35% as being the most likely net increase in the atmosphere as compared with the total output yields 332 ppm for 1980 and 371–378 ppm for year 2000 if using the values for CO_2 output based on the OECD estimates. But a value of 45%, which is also quite possible yields a considerably larger increase (cf. Table 9).

4.2. SEASONAL VARIATIONS

The deduced seasonal variations are very regular and are suitable for some considerations of the vertical exchange processes in the atmosphere. Let us assume that the vertical exchange through the troposphere from 1 km to the tropopause can be described with the aid of a constant eddy exchange coefficient (K), that the observed variations are due to sources and sinks at the surface and that horizontal exchange is not of importance in the present context. If the observed CO_2 variations at the lowest level ($Z_1 = 2$ km) are given as a function of time (t) by

$$q(z_1) = q_0(z_1) + \sum_n A_n \sin\left(\frac{2\pi n}{T} t - \alpha_n\right)$$

Table 10. *Values for the turbulent exchange coefficient, K, as deduced from the change with elevation of amplitude and phase of the annual and semi-annual variation of CO_2*

Parameter used for computation	K ($cm^2\ sec^{-1}$)
A_1	$1.4 \cdot 10^5$
α_1	$1.7 \cdot 10^5$
A_2	$2.6 \cdot 10^5$
α_2	$4 \cdot 10^5$

the variations at the levels above ($z \geq z_1$) will be

$$q(z) = q_0(z) + \sum_n A_n \exp\left[-(z-z_1)\sqrt{\frac{\pi n}{KT}}\right] \sin\left[\frac{2\pi n}{T}t - (z-z_1)\sqrt{\frac{\pi n}{KT}} - \alpha_n\right]$$

The annual increase has been eliminated, $q_0(z)$ is the annual average and T is equal to one year. The amplitude and phase of $q(z) - q_0(z)$ is given at five levels (Table 4). We compute the best fit of these data for the annual and semi-annual wave to a model characterized by constant exponential decrease of amplitude and constant phase shift with altitude as prescribed by the solution above. We then are able to compute four different values for K (cf. Table 10). The amplitude decrease is considerably more accurately determined than the phase shift and the relative error of the semi-annual amplitude is about three times the corresponding error of the annual amplitude. With due regard to these facts we compute that $K = 2 \cdot 10^5$ cm^2 sec^{-1}. As is indicated by the way in which the variability of CO$_2$ in the atmosphere depends on the scale, the intensity of the exchange as expressed by this value for K is due to both synoptic scale, meso-scale and possibly also small scale turbulent processes. The decrease with height of the amplitude of the annual oscillation is more rapid in the lower than in the upper troposphere and a corresponding computation for the layer 6–10 km gives a mean value for $K \approx 6 \cdot 10^5$ cm^2 sec^{-1}.

The annual and semi-annual oscillations decrease rapidly through the tropopause and into the stratosphere. A marked phase shift also occurs. Proceeding in the same way as before, we find a value for the vertical eddy exchange coefficient $K \approx 10^3$ cm^2 sec^{-1} already in the very lowest layer of the stratosphere. This value should, however, not be taken too seriously since the change of amplitude and the phase shift are not determined very accurately. Furthermore Manabe et al. (1965) have shown that horizontal exchange processes between tropical and middle latitudes are of considerable importance for the maintenance of the moisture of the lower parts of the stratosphere. The same may be true with regard to the seasonal variations of the CO$_2$-content of the atmosphere at these levels.

4.3. COMMENTS ON THE USE OF THE CO$_2$ INFRARED EMISSION FOR TEMPERATURE MEASUREMENTS FROM SATELLITES

The successful launch of Nimbus III in April 1969 has shown that the indirect sounding technique for measuring temperature in the troposphere and the stratosphere with the aid of the CO$_2$ emission in the infrared is feasible. The reduction of the radiometer data to information on atmospheric temperature is either based on certain empirical regression formulae utilizing the available radiosond observations or on an inversion of the radiative transfer equation. The present analysis shows that the annual increase and the seasonal variations are quite well known. With the aid of the values in section 3 we are able to compute a representative value for the CO$_2$-content of the air volume from which the radiation is received with an accuracy ±2 ppm in the lower troposphere and ±1.5 ppm in the upper troposphere and lower stratosphere.

REFERENCES

Bischof, W. 1965. Carbon dioxide concentrations in the upper troposphere and lower stratosphere. *Tellus 17,* 397–401.

Bischof, W. 1970. CO$_2$ measurements from air craft. To be published in *Tellus 22,* 5.

Bischof, W. & Bolin, B. 1966. Space and time variations of the CO$_2$ content of the troposphere and the lower stratosphere. *Tellus 18,* 155–159.

Bolin, B. & Eriksson, E. 1959. Changes in the carbon dioxide content of the atmosphere and sea due to fossil fuel combustion. *The Atmosphere and Sea in Motion.* The Rockefeller Institute Press, 130–142.

Bolin, B. & Keeling, C. D. 1963. Large scale atmospheric mixing as deduced from seasonal and meridional variations of carbon dioxide. *J. Geoph. Res. 68,* 3899–3920.

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

Cochrane, J. D. 1958. The frequency distribution of water characteristics in the Pacific Ocean. *Deep-Sea Res. 5,* 111–127.

Craig, H. 1957. The natural distribution of radio carbon and the exchange time of carbon dioxide between the atmosphere and sea. *Tellus 9,* 1–16.

Eriksson, E. 1963: Possible fluctuations in atmospheric carbon dioxide due to changes in the properties of the sea. *J. Geoph. Res. 68*, 3871–3876.

Eriksson, E. & Welander, P. 1956. On a mathematical model of the carbon cycle in nature. *Tellus 8*, 155–175.

Fonselius, S., Koroleff, F. & Wärme, K.-E. 1956. Carbon dioxide variations in the atmosphere. *Tellus 8*, 176–183.

Hutchinson, G. E. 1954. The biochemistry of the terrestrial atmosphere, carbon dioxide and other compounds. In *The Earth as a Planet* (ed. G. P. Kuiper), part II, 375–433. The University of Chicago Press.

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

Lundegårdh, H. 1957. *Klima und Boden in ihrer Wirkung auf das Pflanzenleben.* Gustav Fischer, Jena.

Manabe, S., Smagorinsky, J. & Strickler, R. F. 1965. Simulated climatology of a general circulation model with a hydrological cycle. *Mon. Wea. Rev. 93*, 769–798.

Montgomery, R. B. 1958. Water characteristics of Atlantic Ocean and world ocean. *Deep-Sea Res. 5*, 134–148.

Nydal, R. 1968. Further investigations of the transfer of radio carbon in nature. *J. Geoph. Res. 73*, 3617–3635.

Organization for Economic Cooperation and Development, 1966: Energy policy. Problems and objectives, OECD, Paris.

Pollak, M. J. 1958. Frequency distribution of potential temperatures and salinities in the Indian Ocean. *Deep-Sea Res. 5*, 128–133.

Rafter, T. A. & O'Brien, B. J. 1970. Exchange rates between the atmosphere and the ocean as shown from recent ^{14}C measurements in the South Pacific. *Nobel Symposium*, Stockholm.

Revelle, R. & Suess, H. 1957. Carbon dioxide exchange between atmosphere and ocean and the question of an increase of atmospheric CO_2 during past decades. *Tellus 9*, 18–27.

Rodin, L. E. & Basilevič, N. I. 1968. World distribution of plant biomass. Functioning of terrestrial ecosystems at the primary production level. UNESCO, pp. 45–52.

Young, J. A. & Fairhall, A. W. 1968. Radio carbon from nuclear weapons tests. *J. Geoph. Res. 73*, 1185–1200.

ВАРИАЦИИ СОДЕРЖАНИЯ УГЛЕКИСЛОГО ГАЗА В АТМОСФЕРЕ СЕВЕРНОГО ПОЛУШАРИЯ

Представлены результаты измерений содержания углекислого газа в тропосфере и нижней стратосфере, выполненных в течение шести лет (1963–1968 гг.). Данные показывают годовое увеличение содержания CO_2 в среднем на $(0{,}7 \pm 0{,}1) \cdot 10^{-6}$ в год, тогда как среднее годовое увеличение промышленных выбросов CO_2 составляло приблизительно от $1.9 \cdot 10^{-6}$ до $2.3 \cdot 10^{-6}$ в год. Таким образом, улвеичение в атмосфере составило примерно $\frac{1}{3}$ от общего выброса. Рассмотрение возможного увеличения ассимиляции CO_2 растительностью благодаря его более высокой концентрации показывает, что оно может составлять максимум $\frac{1}{4}$ от полного объема выбросов, вероятно же, значительно меньше. Таким образом, чистый перенос в океаны должен составлять минимум половину промышленных выбросов. Скорость переноса через поверхность океана представляется достаточно эффективной, чтобы преодолеть значительное сопротивление растворению, и решающим фактором в определении скорости переноса поэтому является циркуляция океана или скорость его перемешивания. Полученные цифры указывают, что примерно 20–25% массы воды в океане должны быть доступны для растворения в течение времени быстрого увеличения индустриальных выбросов CO_2 (за последние 30–50 лет), чтобы объяснить извлечение из атмосферы довольно больших количеств CO_2. Продолжающееся увеличение сжигания ископаемых топлив предполагает, как это предсказывается ОЭКР (Организацией для экономической кооперации и развития), что содержание CO_2 в конце столетия будет между $370 \cdot 10^{-6}$ и $395 \cdot 10^{-6}$, по сравнению с $320 \cdot 10^{-6}$ — средней цифрой для 1968 г. Найдено, что амплитуда сезонных изменений порядка $6{,}5 \cdot 10^{-6}$ на высоте 2 КМ и $3{,}5 \cdot 10^{-6}$ в верхней части тропосферы. Фазовый сдвиг сезонных вариаций между этими двумя уровнями порядка 25–30 дней. На основе этих данных получено, что коэффициент вертикального турбулентного перемешивания $K = 2 \cdot 10^5 \text{см}^2\,\text{сек}^{-1}$. Амплитуда сезонных вариаций в нижней стратосфере на высотах 11–12 КМ меньше $1 \cdot 10^{-6}$ и имеет отставание по фазе, по крайней мере, на полтора месяца по сравнению с верхней тропосферой.