Variation in C^{14} concentration in the atmosphere during the last several years

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ABSTRACT

Variation in C^{14} concentration with time at various northern latitudes from 9°N to 78°N has been studied in the troposphere mainly since 1963 (NYDAL, 1963, NYDAL & LÖVSETH, 1965). Sampling has later been extended to 21°S. During 1965 sampling was also performed at 63°N at an altitude of 13 km.

Our tropospheric data has been compared with the HASL measurements (HAGEMANN *et al.*, 1965) at an altitude of 20 km. It appears that the stratosphere at this height mixes very rapidly in the latitudinal direction at latitudes above 30°N. The diffusion coefficient for turbulent mixing in this region may be of the same order as that of the troposphere (10^{10}–10^{11} cm²/sec., MÜNNICH, 1963).

In 1963 there seems to be a decrease in exchange rate below about 30°N, both within the stratosphere (20 km altitude) and within the troposphere. In summer 1964, however, this "barriere" in the troposphere is less convincing because all the C^{14} peaks occur nearly simultaneously from 9°N to 78°N. The peak at Dakar (15°N) is even higher than several peaks farther north.

The author's impression is that the simultaneous occurring C^{14} peaks in the troposphere in summer at various northern latitudes are partly due to a simultaneous removal of C^{14} from the stratosphere to the troposphere at the respective latitudes. It turns out that C^{14} from polar explosions in 1962 is transferred to the lower and southern latitudes both through the troposphere and through the stratosphere. C^{14} transfer through the last one seems to be important.

Introduction

C^{14} produced in nuclear explosions has been a useful tracer for studying transport processes in the atmosphere. During the last 10 years, studies of mixing in the troposphere, mixing across the tropopause and within the stratosphere have been the objects for several investigators.

The hydrogen bomb provides a larger production of C^{14} per megaton than the former bombs, and the C^{14} isotope has, therefore, during the last years been a more useful atmospheric tracer than before.

Our interest for C^{14} in atmospheric CO_2 was caught on a rather late stage. The occasion for doing the present measurements was the large series of thermonuclear tests, which were carried out in 1961 and 1962 mainly above the Barents Sea between 70°N and 80°N. One of the purposes of our measurements was to examine transport processes in the atmosphere. We were especially looking for latitudinal variation in C^{14} in the northern troposphere. We believed that there would be a much larger descending of C^{14} from the stratosphere in the polar region than farther south, and therefore a latitudinal effect in the troposphere should be expected.

1. Sampling and localities

The sampling procedure was the same as introduced by RAFTER & FERGUSSON (1957), and consisted of a sodium hydroxide solution placed in open air. We used a 2 liter 1/2 N solution placed in a dish of 450 cm² cross section, and exposed over 7 days. The amount of CO_2 absorbed during this time was normally 3–5 litres. The water applied in the solution was either rain water or distilled water. During winter time on higher latitudes (above 58°N), a

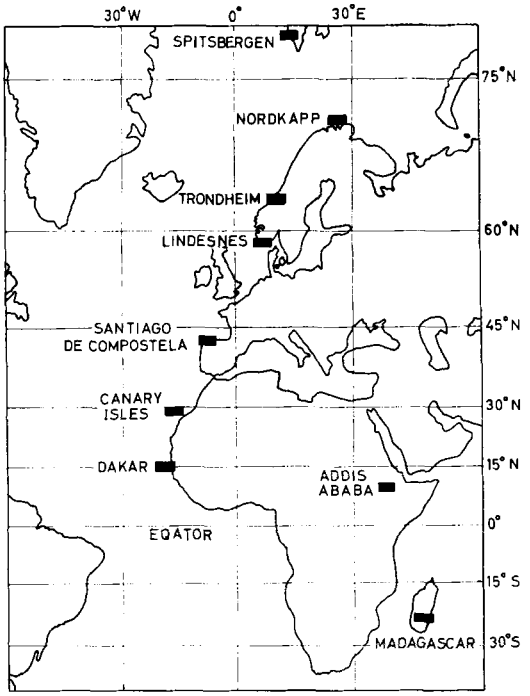

FIG. 1. CO_2-sampling stations.

thermostatically controlled heating element of 40 W in the solution, served to keep the temperature slightly above zero.

The sampling localities were chosen with the main purpose of obtaining the most representative C^{14} value in the troposphere at each latitude. In addition to sampling along the coast of the North Atlantic Ocean from 15°N to 78°N (Fig. 1), samples were also collected at Addis Ababa (9°N), and later in Madagascar (21°S). The distance between each sampling station was partly determined from earlier experiences by other laboratories (MÜNNICH, 1963), and partly it was a practical problem to have good sampling stations established.

Spitsbergen: The samples were collected by the people of the radio- and meteorological station at Kapp Linné (78°04′N, 13°38′E). The head of this station was Kåre Henriksen until July 1964, followed by Ingvar Johansen. This station belongs to Telegrafstyret in Oslo. The absorption dish was placed about 10 m above sealevel, and at least 50 m away from the chimney.

Nordkapp: The samples were collected by the head of Fruholmen Lighthouse (71°06′N, 23°59′E). Arild Klefstad was in charge of this work until July 1964, followed by Odd Salomonsen. The absorption dish was placed on the platform in the tower of the lighthouse, 70 m above sealevel. The lighthouse is electrically operated from a power station on the main land.

Trondheim: The samples were collected by the staff of the Radiological Dating Laboratory. In 1962 collection took place on a hill (63°25′N, 10°15′E), 450 m above sealevel, 6 km away from the town. When the activity began to decrease in winter time, we were afraid of contamination of CO_2 from fuel combustion in the town. Therefore, from the beginning of 1963 the sampling was continued at a different locality (63°16′N, 10°22′E), 700 m above sealevel, 20 km away from the town. No difference in activity, however, was observed by this change.

Lindesnes: The samples were collected by the head of Lindesnes Lighthouse (57°59′N, 07° 04′E) Reinert Rasmussen until November 1963, followed by Ingvar Johansen. The absorption dish was placed on a platform in the tower of the lighthouse, 50 m above sealevel. The lighthouse is electrically operated from a power station far away.

Santiago de Compostela: The samples were collected by the meteorologist Enrique Lazo Alcalá del Olmo, Aeropuerto, Santiago de Compostela (42°53′N, 08°26′W). The absorption dish was placed 1.5 m above the ground in the corner of the airport, situated 360 m above sealevel, 10 km away from the town. The main wind direction at the absorption dish is away from the runway, 600 m apart.

Gran Canaria: The samples were collected by C. A. Rouiller, Jr., director of the N.A.S.A. Tracking Station (27°30′N, 15°30′W). The absorption dish was placed close to the shore, 10 m above sealevel.

Tenerife: The samples were collected by director Fransisco Sánchez Martinez, and meteorologist Fernando Molina Herrero, Observatorio Astronomico, Izana (28°22′N, 16°30′W). The absorption dish was placed about 100 m away from the observatory, which is situated 2400 m above sealevel in the slope of Pico del Teide.

Dakar: The samples were collected by Drs. P. Elouard and B. Diagne, Université Dakar-Fann, Sénégal. Until the autumn of 1964, collection took

place at the shore (14°33′N, 17°07′W), 10 m above sealevel, 70 km away from the town. The samples were later collected on the roof of the Geological Department, Université Dakar-Fann (14°41′N, 17°28′W). This institute is situated on a little isle. Simultaneous measurements on the 2 localites gave no important difference in activity.

Addis Ababa: The samples were collected by the teachers Hadle Hovda and Oskar Nydal, Ethiopian Evangelical College, Debre Zeit (8°40′N, 38°58′E), 1900 m above sealevel. The absorption dish was placed on the roof of a building.

Madagascar: The samples were collected by the Norwegian missionary Earl Lohne, Fianarantsoa (21°27′S, 47°05′E), 1100 m above sealevel. The absorption dish was placed 3 m above the ground.

High altitude sampling: High altitude sampling was started in April 1965 in the vicinity of Trondheim (63°N) at about 13 km altitude. The sampling was carried out by Colonel Lieutenant Knud Lundell and Captain Stener Hveem of the Norwegian Air Force.

CO_2 was collected in a filter container placed under the wing of the plane. The filter contained about 1,4 kg molecular sieve material of the type 4A (FERGUSSON, 1963), which was exposed for 15 min. The 2 shutters, one in each end of the filter container, were electrically driven and operated by the pilot.

2. Chemical treatment and measurements

The exposed solutions contained dissolved Na_2CO_3 and HCO_3. For most of the time a precipitation to $CaCO_3$ with $CaCl_2$ has been performed. The $CaCO_3$ was further washed and dried and transferred under vacuum to CO_2 by treatment with hydrocloric acid. Afterwards the CO_2-gas passed a purification system, which removed electronegative impurities. It later became apparent that the exposed solutions could be directly treated with hydrocloric acid under vacuum. The CO_2-gas produced in this way became extremely pure after having passed through the ordinary purification system. The latter treatment of the samples had the advantage that no radon was introduced in the CO_2-gas.

The exposed molecular sieve material was treated in an iron furnace at a temperature of 500°C. At this temperature the absorbed CO_2-gas was removed in 4–5 hours.

The C^{14} measurements are mainly carried out in 2 CO_2 proportional counters with effective volumes of 1.1 and 1.5 litres. The counters are working at a pressure of 2 atmospheres CO_2. The backgrounds of the counters are respectively 3.0 and 2.0 c/min, with corresponding recent standard net counts of 14.2 c/min and 19.2 c/min.

The increase in C^{14} activity is calculated in per cent excess above natural C^{14} level. This level represents the normal C^{14} activity in recent wood in absence of man-made C^{14}. The N.B.S. oxalic acid standard (BROECKER & OLSON, 1959) has been applied in our calculation.

The C^{13}/C^{12} ratio has been mass-spectrometrically measured in most of the samples in order to study fractionation between the carbon isotopes absorbed in the solution. It appeared that the C^{13}/C^{12} ratio was very close to that of recent wood. The corrected C^{14} excess (ΔC^{14}) is calculated according to the formula

$$\Delta C^{14} = \delta C^{14} - (2\delta C^{13} + 50)\left(1 + \frac{C^{14}}{1000}\right)$$

where δC^{14} represents the uncorrected C^{14} excess in percent relative to the N.B.S. standard, and δC^{13} represents the deviation in C^{13}/C^{12} ratio relative to the P.D.B. Standard (CRAIG, 1961). The limit of error (one standard deviation) in each ΔC^{14} value is generally between 1.0 and 1.5 %, and includes only the statistical error in the counting result and the error in the C^{13}/C^{12} ratio. The latter is measured with an accuracy of 0.06 per cent at Karolinska Institutet, Stockholm. For the stratospheric samples, however, the error is appreciably greater and may constitute values up to 10 %. This error (probable error) is largely caused by contamination with tropospheric CO_2, because of a leakage in the valves of the sample container.

3. Results and discussion

58°N–78°N (Fig. 2)

The increasing part of the curves in spring and summer 1963 is equal to the above 4 localities. The curves simultaneously reach the same height of about 100 % above natural

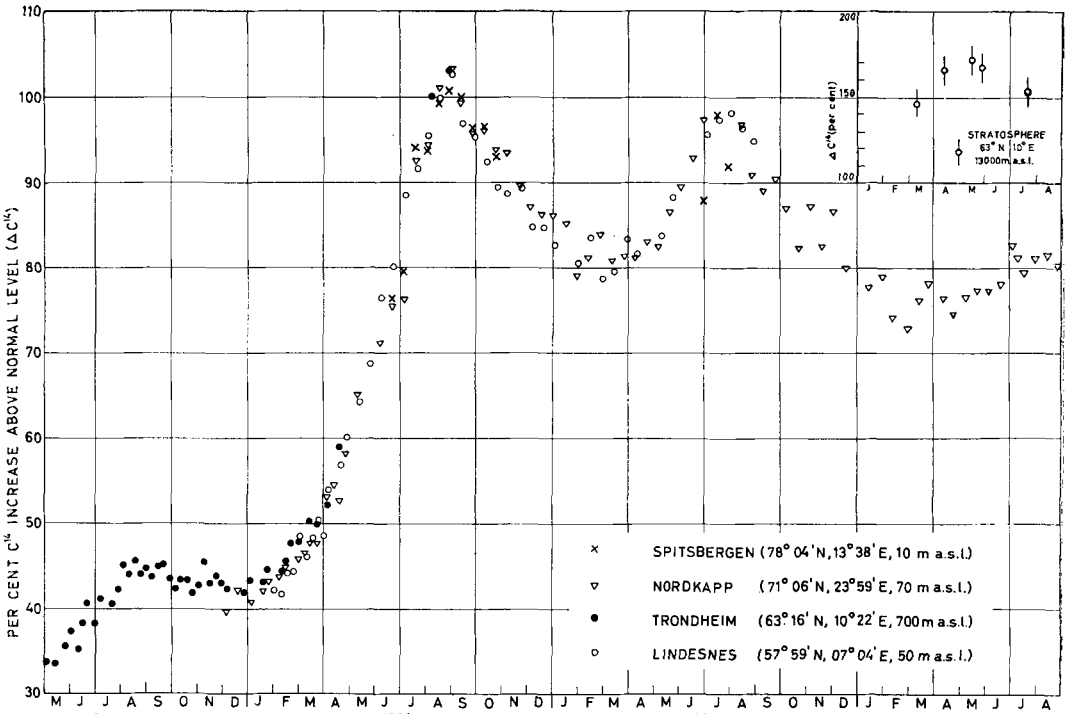

FIG. 2. Variation in C^{14} between 58°N and 78°N.

level at September 1. All the curves decline in approximately the same manner during winter time. There may be a weak tendency of a more rapid decrease at 58°N than at 71°N to 78°N. In August 1964 the curves at 58°N and 71°N reached a height of 98 %. The peak at 71°N in 1965 was only 82 % above natural level. It turns out that the maximum peak was already reached in 1963, and all the prospective peaks will successively become smaller.

Because of the equality in the observations between 58°N and 78°N, the number of samples measured in this region has gradually been reduced. Measurements at Trondheim were stopped in September 1963, and at Spitsbergen only a few of the collected samples have been measured. For several years, regular measurements at Spitsbergen (78°N) and Abisko (68°N) have been carried out by the laboratory in Uppsala (OLSSON & KARLÉN, 1965). These measurements agree well with our own and confirm the opinion that there is no important difference in C^{14} variation in the region 58°N to 78°N. There is also good agreement between the C^{13}/C^{12} values in the above region. The deviation relative to the P.D.B. standard constitutes values between −23 and −26 per mill. There is a maximum enrichment in C^{13} in July–August 1963, and a minimum in February–March 1964 (Fig. 5). KEELING (1958), who has observed a similar variation, suggests that it may be caused by the biological cycle of the terrestrial biosphere. The normal δC^{13} value in the atmosphere at the above latitudes is about −7 ‰. Even with some fluctuation in the fractionation by absorption, it is suggested that the observed mean variation in the C^{13}/C^{12} ratio reflects a true variation in the troposphere.

27°N – 43°N (Fig. 3)

It was observed that the increase in activity during spring 1963 is equal and simultaneous at Gran Canaria, Santiago de Compostela and the northernmost latitudes. The peak activities reach nearly the same height at all localities, but the peak on Gran Canaria was delayed relative to the other peaks. This delay was maintained in the declining part of the curve throughout the year. The C^{14} activity on

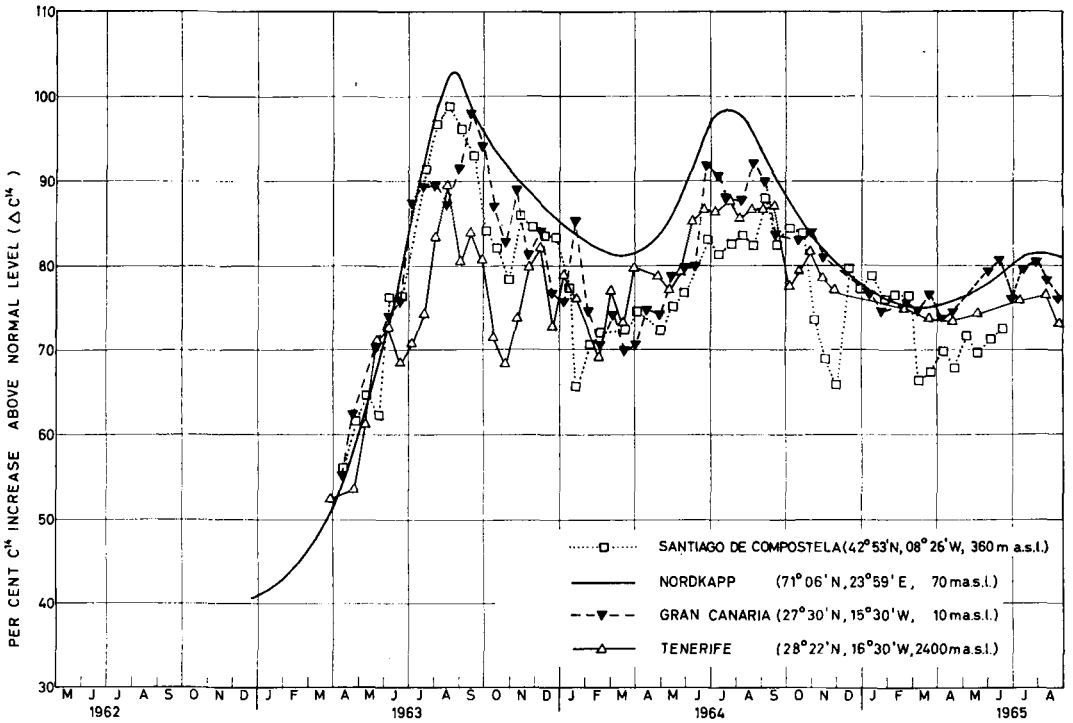

FIG. 3. Variation in C^{14} between 27°N and 43°N.

Tenerife shows in the period July to December 1963 an appreciable deviation (up to 10 %) from the activity observed farther north. In summer 1964 Tenerife has a higher C^{14} activity than Santiago de Compostela, but a lower activity than Gran Canaria. The prevailing wind direction at sealevel on Gran Canaria throughout the year is SW, and NW on Tenerife at a height of 2400 m. It seems reasonable that the sampling stations on respectively Gran Canaria and Tenerife sometimes are receiving air belonging to separate air masses, which mixes in a relatively long time. Meaurements of the C^{13}/C^{12} ratio seem to some extent to state this assumption (Fig. 5). It is apparent that in the period August to November 1963 the samples on Gran Canaria and Santiago de Compostela are more enriched in C^{13} and C^{14} than was the case on Tenerife. During the remaining part of 1963 and throughout 1963 the δC^{13} values on Tenerife and Santiago de Compostela are in good agreement (21–24 ‰), whereas the δC^{13} values from samples on Gran Canaria are often irregular and show large deviation (5 ‰) from the others, especially in summer 1964.

The main trend, however, is that the C^{14} curves on various latitudes are very similar close down to Gran Canaria. This is also in agreement with other measurements (LAL & RAMA, 1966). Most of the small deviations between the curves may not be important from a meteorological point of view. Such deviations may change from year to year depending on varying meteorological conditions.

9°N–15°N (Fig. 4)

In 1963 the C^{14} activity at Dakar (15°N) and Addis Ababa (9°N) seems to have a clear deviation from any curve farther north in the way that the large summer peak is vanishing. The activities at both localities are increasing throughout 1963. In 1964 there is a large peak at Dakar with a maximum height of 80 % above natural level. This peak occurs simultaneously with the peaks farther north and is even a few percent higher than the peak on Tenerife and Santiago de Compostela. The activity at Addis Ababa shows a systematic lower concentration than at Dakar. It is not quite clear if the last difference is mainly a lati-

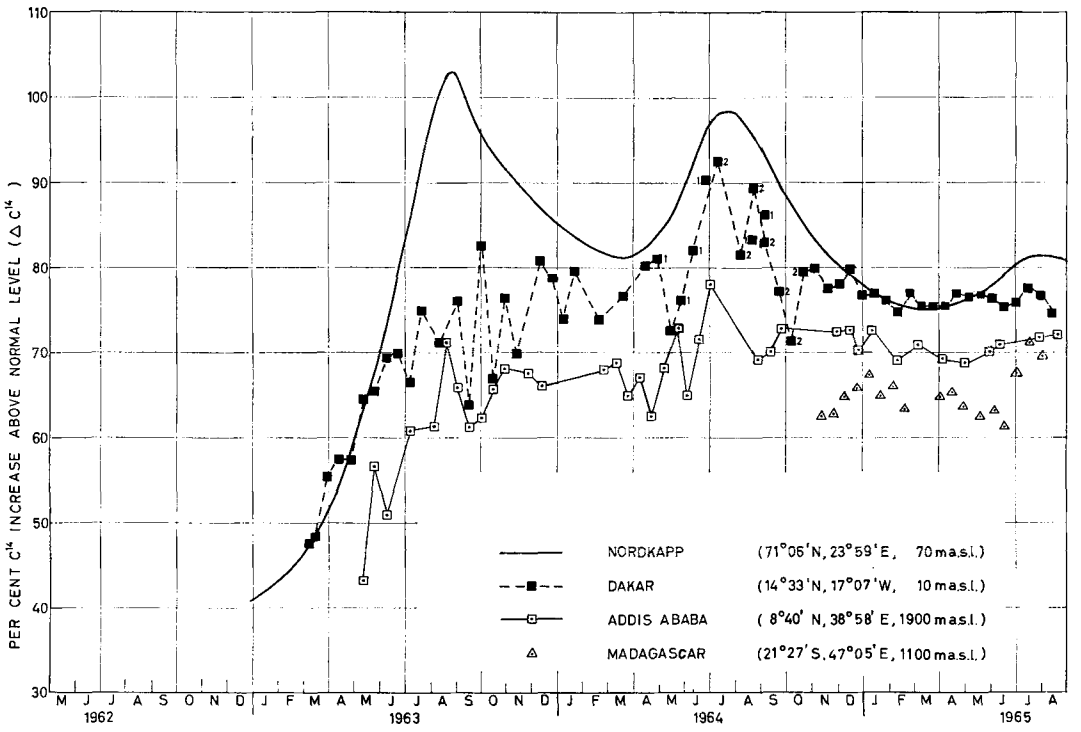

FIG. 4. Variation in C^{14} between 21°S and 15°N.

tudinal effect or an effect due to different altitudes. There tends to be a small summer peak at Addis Ababa both in 1963 and 1964. These peaks are simultaneous with the peaks farther north. It turns out that the C^{14} activity from Addis Ababa and Dakar is nearly representative for the prevailing activity at the respective latitudes around the world (LAL & RAMA, 1966).

The δC^{13} values of the samples from Dakar and Addis Ababa are mainly found in the region -20 to -23 ‰. Compared with the values observed farther north, there tends to be an increasing enrichment in C^{13} with lower latitudes. There also tends to be a seasonal variation in the C^{13}/C^{12} ratio in the tropic.

21°S (Fig. 4)

Measurements in Madagascar have only been carried out since November 1964. Other measurements taken between 18°S and 41°S (Lal and Rama, 1966) show that there has been a nearly linear increase in C^{14} in the southern troposphere during 1963 and 1964. Our measurements from 21°S show some variation in the activity. The data are, however, too few in order to make any definite conclusion about the correspondance in C^{14} variation between the northern and southern troposphere.

As regards the C^{13}/C^{12} measurements from 21°S, the δC^{13} values obtained so far are between -17 and -20 ‰, and consequently lower than the values found on the other side of Equator.

High altitude sampling (Fig. 2)

Sampling at about 13 km altitude, 63°N, has been carried out only during spring and summer 1965. In August 1965 the C^{14} excess in the lower stratosphere was found to be only about twice that of the troposphere.

It was of great value to study the HASL measurements (HAGEMANN *et al.*, 1965) in the stratosphere during 1963 and 1964. In the region between 34°N and 71°N at 20 km altitude, the stratosphere seems to be mixed very rapidly. The diffusion coefficient for turbulent mixing may be of the order of 10^{10}–10^{11} cm²/sec. The C^{14} concentration at 20 km altitude in the above mentioned band was 17 to 19

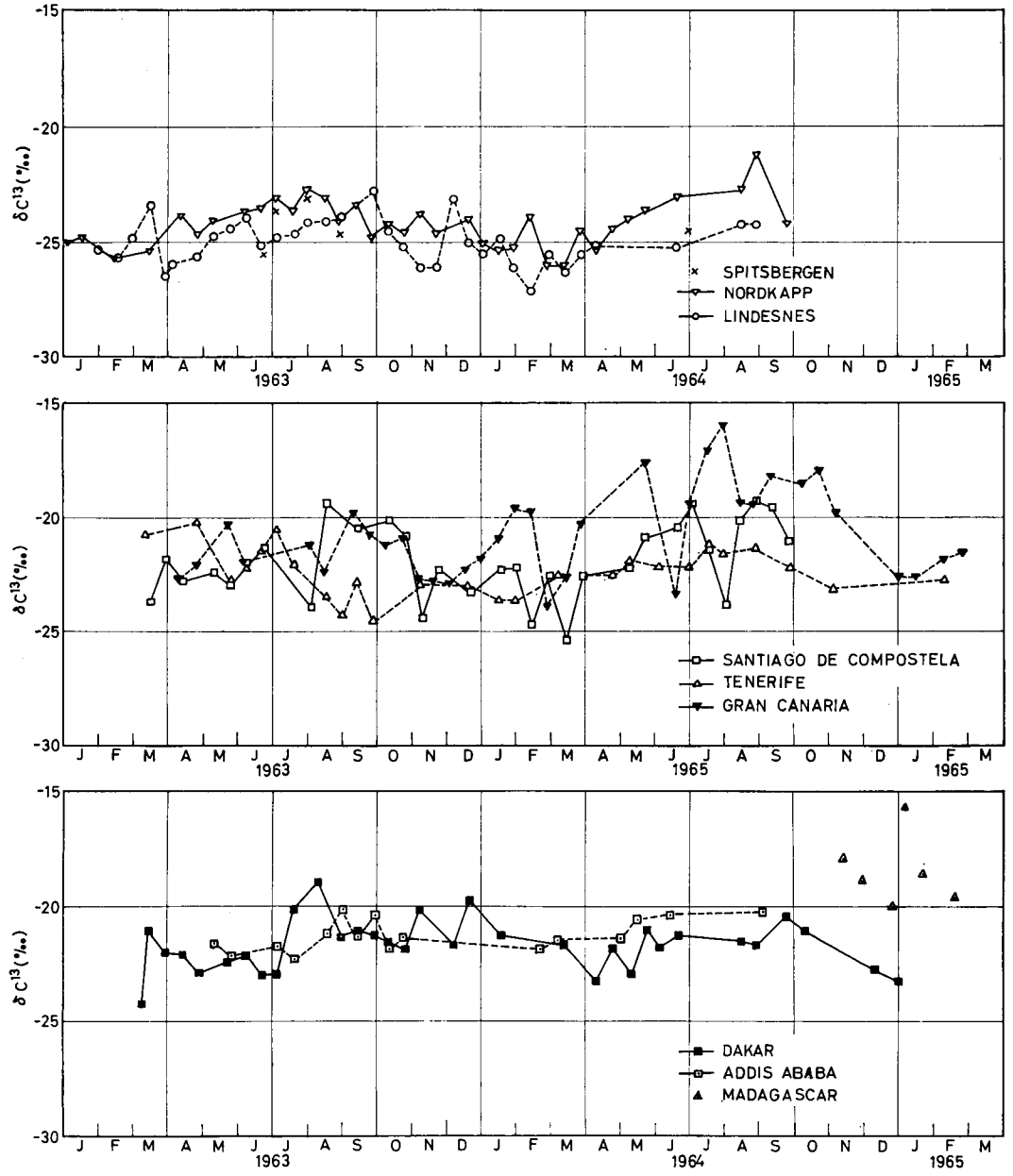

FIG. 5. C^{13}/C^{12} variation at various latitudes.

times the normal value in July–August 1963. The decrease seems to be nearly linear the following year, and the activity was reduced to 7–9 times the normal in July–August 1964.

In the region 10°N to 31°N at 20 km altitude, there seems to be a marked decrease in exchange rate from that observed at latitudes farther north. In July–August 1963 the C^{14} concentration at 10°N was 7–8 times the normal, and in July–August 1964 it was 4–5 times the normal. In the southern stratosphere at 20 km altitude and 42°S, the C^{14} concentration was 2–3 times the normal throughout 1963. According to the HASL measurements, the half

removal time of C^{14} at 20 km altitude seems to be about 1 year at various northern latitude. This is also in agreement with earlier measurements (Rep. U.N., 1962).

4. Concluding remarks

There seems to be a nearly uniform concentration of C^{14} in the stratosphere from early 1963 in the region 34°N to 71°N at 20 km altitude. This is also the case in the troposphere in the region from about 28°N to 78°N. The exchange rate both within the stratosphere (20 km altitude) and the troposphere seems to decrease with lower latitudes. In 1963 the main step in the exchange rate seems to occur in the vicinity of 30°N. This is also verified with tritium and strontium measurements in the troposphere (LAL & RAMA, 1966). The simultaneously occurring summer peaks in the northern troposphere seem to some extent to be independent of the latitudinal exchange rate in the troposphere. The peaks seem to be more a result of a simultaneous removal of C^{14} from the stratosphere at various latitudes, rather than a rapid mixing in the troposphere of locally descended C^{14} from the stratosphere. In 1964 all peaks occur nearly simultaneously from 9°N to 78°N, and the peak at Dakar (15°N) is even higher than several peaks farther north. C^{14} from polar explosions is transferred to the lower and southern latitudes both through the stratosphere and the troposphere. For the large bombs used in 1962 the transport through the stratosphere is probably of great importance.

Taking into account all C^{14} measurements in both the northern and southern troposphere, it appears that the mean maximum C^{14} level in the whole troposphere was already reached in 1964 and constituted an excess of about 75 % above natural level. The prospective decrease in tropospheric C^{14} is mainly determined by the C^{14} excess in the stratosphere and the exchange rate of CO_2 between the troposphere and the stratosphere, and between the troposphere and the ocean. The exchange rate of CO_2 between the troposphere and the surface layer of the ocean is not well established. The mean residence time for a CO_2 molecule in the troposphere before it is absorbed in the ocean is estimated to range from 1.4 years (RAFTER & FERGUSSON, 1958) to 7 years (MÜNNICH & VOGEL, 1958). If no further nuclear tests are carried out the next 1 or 2 years, there will be an excellent opportunity to study the exchange rate between the troposphere and the surface layer of the ocean.

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ИЗМЕНЕНИЯ КОНЦЕНТРАЦИИ C^{14} В АТМОСФЕРЕ В ТЕЧЕНИЕ ПОСЛЕДНИХ НЕСКОЛЬКИХ ЛЕТ

С 1963 года изучалось изменение со временем *концентрации* C^{14} *в тропосфере на различных* широтах от 9° до 78° с. ш. (Nydal 1963, Nydal и Löfseth 1965. Позднее взятие проб воздуха было распространено до 21° ю. ш. В течение 1965 года пробы также брались на высоте 13 км при широте 63° с. ш.

Наши тропосферные данные были сравнены с измерениями HASL на высоте 20 км (Hagemann и др. 1965 г.). Оказывается, что в стратосфере на этих высотах происходит очень быстрое перемешивание в широтном направлении для широт больших 30° с. ш. Коэффициент диффузии для турбулентного перемешивания в этой области возможно имеет тот же порядок, что и в тропосфере (10^{10}–10^{11} см²/сек, Munnich 1963).

В 1963 году казалось имеет место уменьшение скорости обмена как в стратосфере *(на высотах 20 км), так и в тропосфере ниже* приблизительно 30° с. ш. Однако летом 1964 г. эти «барьеры» в тропосфере было менее убедительные, потому что все пики в концентрации C^{14} имели место почти одновременно от 9° до 78° с. ш. Пик в Дакач'е (15° с. ш.) был даже выше, чем отдельные пики севернее.

По впечатлению автора, указанные пики в тропосфере частично вызваны перемещением C^{14} из стратосферы в тропосферу на соответственных широтах. Оказывается, что из полярных взрывов в 1962 г. C^{14} переносился в более низкие северные и южные широты, как через тропосферу, так и стратосферу, причем перенос через последнюю по всей видимости является более важным.