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The Relationship between the Atmospheric Carbon Dioxide Concentration and the Carbon-14 Activity in Tokyo

Sakuji IKEDA, Hideki SHIRAKAWA, and Iwakazu HATTORI

Research Laboratory of Resources Utilization, Tokyo Institute of Technology, Meguro-ku, Tokyo 152

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Synopsis. Measurements of the atmospheric CO_2 concentration and the ^{14}C activity in Tokyo were carried out. The activity of ^{14}C in the CO_2 decreased with an increase in the atmospheric CO_2 concentration, indicating that increases in the atmospheric CO_2 are caused only by the burning of fossil fuels.

Studies of atmospheric CO_2 have been carried out either by the measurement of the concentrations or by ^{14}C-activity measurement. Thus, measurable increases on a global scale have been reported for the atmospheric CO_2 concentrations at several locations; thus, the increases are not the result of local influences.[1] An alternative evidence for the global increases caused by the burning of carbonaceous fossil fuels is the Suess effect,[2] which is the decrease in atmospheric ^{14}C concentrations caused by the combustion of ^{14}C-free fossil fuels.

Since the increments in the atmospheric carbon dioxide concentration at any site reflect the emissions of CO_2 from man-made combustion sources as well as from natural sources, it is not possible to identify the type of source only by the concentration measurement; simultaneous measurements with the ^{14}C activity in the atmospheric CO_2 are necessary to identify the type of source and to quantify the fossil CO_2 in the atmosphere. The source analysis in the atmospheric CO_2 would be of value in studies not only of air pollution but also of the circulation of carbon and of climatic influence.

Toward this purpose, we have been conducting measurements of atmospheric CO_2 and ^{14}C concentrations in Tokyo since April, 1975. This note is a brief report on the atmospheric CO_2 based on one year of measurement.

Experimental

The concentration of atmospheric CO_2 was continuously recorded by means of a Shimadzu Model URA-2S nondispersive infrared CO_2 analyzer at the Ookayama campus of this Institute (35°35′54″ N, 135°42′18″ E). The sampling probe was set on the roof of the building of this Research Laboratory about 20 m above the ground. The CO_2 was collected two or three times a month. Each time about one mole of CO_2 was collected as sodium carbonate by bubbling air, at the rate of 50 l/min for 24 h, through 3 l of a carbonate-free solution containing 5 mol of sodium hydroxide. The collection was almost quantitative. The carbonate thus obtained was then converted into benzene according to the benzene-radiocarbon-dating process.[3] The samples were counted in a modified Packard Tri-Carb Model 3390 liquid scintillation spectrometer. The activity of ^{14}C was given as the disintegration per minute per gram carbon (dpm/g-C). Calibration curves for the correction of the counting efficiencies of the samples were made by the use of a series of unquenched and least-quenched 10^5 dpm standards prepared from toluene-^{14}C of $5.92 \times 10^5 \pm 3\%$ dpm/g (Packard Instrument Co., Inc.). The standard deviations quoted (1σ) describe the uncertainties associated with toluene-^{14}C, standard preparation, and measurements of the sample, the standard, and the background. The activity of the NBS oxalic acid reference standard was 14.39 ± 0.46 dpm/g-C.

Results and Discussion

Figure 1 indicates the mean monthly CO_2 concentrations from April, 1975, through March, 1976. The maximum and minimum mean hourly concentrations observed through the month are also illustrated in the figure. The seasonal variation is obvious. Since the variation is negatively correlated with the air temperature in Tokyo, since the estimated amount of total fossil fuel consumption, including mobile fuels, in Tokyo during the winter season is double that in summer, and since an extremely high concentration is observed during the winter season, especially at low wind speeds, it can be considered that the principal origin of this variation

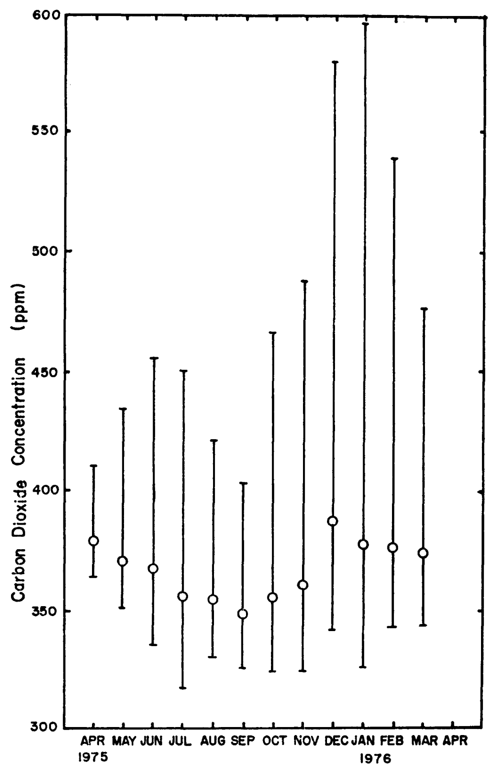

Fig. 1. Mean monthly atmospheric CO_2 concentrations in Tokyo.
Open circles represent mean monthly value. The upper and the bottom limits indicate the maximum and the minimum mean hourly values, respectively, observed through the month.

is due to the combustion of fuels for heating purposes.

The CO_2 concentrations generally decreased with an increase in the wind speed, but never fell for below the values of 317—325 ppm, which can be regarded as the background concentration in air coming into Tokyo. The atmospheric ^{14}C activities (δ ^{14}C % above the modern reference) in 1970 were reported to be 50—59% at Monaco,[4] 44.7—56.4% at Taipei, Taiwan,[5] and 50.4—53.1% at Trnava, Czechoslovakia.[6] The scattered values would appear to indicate a local Suess effect, although the investigations stated that the sampling sites were far from local pollution. Taking into consideration the recent trend of atmospheric ^{14}C activities to decrease as a result of various mixing processes of ^{14}C produced by nuclear weapons testing, coupled with the global Suess effect, the ^{14}C activity in the background CO_2 in 1975 was estimated to be 50.0% above the modern reference or 20.5 dpm/g-C.

The relationship between the atmospheric CO_2 concentrations and the ^{14}C activities is shown in Fig. 2. The smooth curve in the figure represents the calculated relationship under the assumptions that the CO_2 increments from the background concentration are caused only by the fossil CO_2, and that the background concentration and ^{14}C activity in the CO_2 are 325 ppm and 20.5 dpm/g-C respectively. The overall agreement between the observed values and the smooth curve strongly suggests that increases in the atmospheric CO_2 concentration in Tokyo are caused only by the burning of fossil fuels. In fact, the estimated amount of fossil CO_2 released into atmosphere over Tokyo in 1975 reached 7.5×10^{13} g, whereas natural emissions from green areas in Tokyo were estimated to be less than 1.8×10^{11} g/year. On the other hand, the CO_2 from human resipiration and from the decomposition of human excreta can be calculated to be 0.42×10^{13} g/year based on the population in Tokyo and the amount of food consumed. The activity of the ^{14}C in the CO_2, of course, is almost the same as that in natural CO_2.

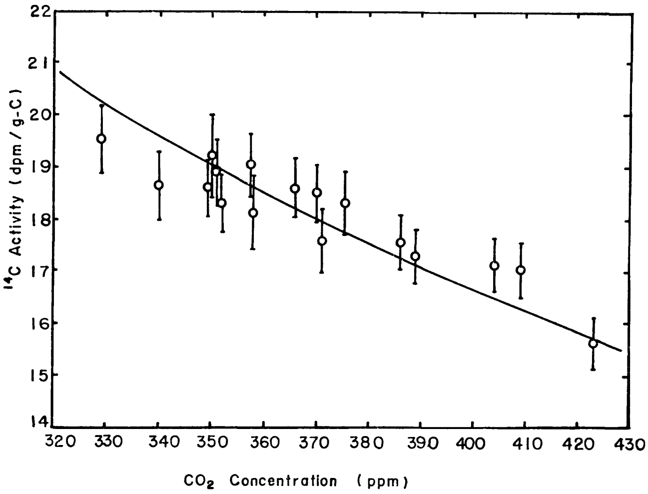

Fig. 2. Relationship between atmospheric CO_2 concentration and ^{14}C activity.
The solid curve represents the calculated relationship by the fossil CO_2.

An analysis with increased precision could be achieved by measurements of the background levels of both the atmospheric CO_2 concentration and the ^{14}C activity in air coming into the observed site, because the background concentrations of atmospheric CO_2 show seasonal variation and have increased at least 1.0 ppm per year after 1969,[1] fluctuations of atmospheric ^{14}C activity as a result of causes have also been observed.[7]

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