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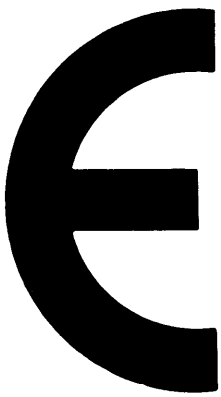

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INVESTIGATIONS INTO THE EMISSION OF CARBON-14 COMPOUNDS FROM NUCLEAR FACILITIES, ITS MEASUREMENT AND THE RADIATION EXPOSURE RESULTING FROM THE EMISSION

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REPORT

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C O N T E N T S

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NOTE: In this report the Continental usage of a comma to indicate a decimal point is followed.

Except where otherwise the use of the word "Federal" refers to the Federal Republic of Germany as in "Federal Minister of the Interior" (page 5). The terms "dose equivalent" and "dose equivalent commitment" have been abbreviated the "dose" and "dose commitment" respectively.

Abbreviations: BWR = boiling-water reactor
PWR = pressurized-water reactor.

Summary

This report deals with the emission of carbon-14 from nuclear facilities (nuclear power plants and reprocessing facilities) and the resulting radiation exposure. A preliminary survey is made of studies concerning the calculation and assessment of the rates of formation and release of C-14 from reactors and reprocessing facilities and previous measurements of emission rates from such facilities are summarized. The methods adopted for sampling and measuring C-14 compounds in air are described, and the results of more than 700 individual measurements for light-water cooled reactors are reported. The results of calculations of the radiation exposure of the population are also discussed.

Earlier studies, as well as the results reported here, validate the emission rates calculated on the basis of nuclear data for boiling-water reactors (BWRs) of 14 Ci/GWa electric power generation, for pressurized-water reactors (PWRs) of 6 Ci/GWa, and for the reprocessing of fuel from light-water cooled reactors of about 13 Ci/GWa.

Individual results from measurements, taken over a period of more than 2 years, of C-14 activity-concentrations in effluent air samples lie in the range of 9 to 24 Ci/GWa for BWRs and 2 to 12 Ci/GWa for PWRs.

With BWRs and the reprocessing plant (purex process) at Karlsruhe the C-14 is emitted almost exclusively as carbon dioxide (> 95% of the total emission). PWRs emit primarily chemical compounds such as carbon monoxide, methane or other hydrocarbon compounds. On average the carbon dioxide component amounts to about 30% for PWRs, but only to about 10% for newer and larger facilities.

For an emission value of 10 Ci/a of CO_2-bound C-14 from BWRs, under standard propagation conditions the maximum local radiation exposure is calculated to be 0,2 mrem/a for bone via ingestion. This calculation is based on the assumption that

vegetables, milk and meat produced exclusively at the point of maximum exposure are ingested. On the other hand, for an emission of 5 Ci/a of C-14 from PWRs with only 10% of the C-14 CO_2-bound, the resulting maximum radiation exposure is 0,01 mrem/a.

Emission of C-14 is also of significance because of the global effects possible throughout the period the C-14 persists in the biosphere. The emission into the atmosphere of 1 Ci of C-14 (in whatever chemical form) causes a collective dose commitment of about 500 man.rem.

An assessment of the individual cancer risk from C-14 shows that the contribution of the C-14 released from nuclear facilities is slight compared with that of the C-14 produced naturally or by nuclear explosions.

When calculating the collective dose commitment, it should be borne in mind that, with the adoption in future reprocessing facilities of techniques for the retention of CO_2-bound C-14, a considerable reduction in the global dose commitment of C-14 is to be expected.

1. Introduction

That carbon-14 (C-14) is formed due to various activation processes in the neutron field of a nuclear reactor has been known for a long time. In a nuclear-engineering publication in 1963 (RAB 63), for example, it was pointed out that radiological protection must take into account the emission of C-14 from reactors. In a publication in 1968 (POV 68) the possibility was expressed that an increased concentration of C-14 could be present in the atmosphere near nuclear reactors.

Measurements of C-14 in the vicinity of nuclear power plants were made as early as 1969 within the scope of an investigation by the U.S. Public Health Service on surveillance of radioactive material from nuclear power plants (KAH 74). Further measurements in the following year were carried out by the radiological protection laboratories of the Health Authorities of New York State (MAT 73). For example, Kunz (KUN 74, KUN 75) determined an emission of about 16 Ci/GWa for boiling-water reactors (BWRs), and of about 6 Ci/GWa for pressurized-water reactors (PWRs). The C-14 appeared in the form of methane (CH_4) and other hydrocarbons or as carbon monoxide (CO) if reducing conditions existed in the cooling medium, or in the form of carbon dioxide (CO_2) if oxidizing conditions prevailed. Since hydrogen is added to the primary cooling material in PWRs, the formation of CH_4 and other hydrocarbons is certainly to be expected with them.

A report was given at the 1974 Reactor Conference of the German Atomic Forum (BON 74) on the rates of formation of C-14 in nuclear fuels and cooling media as calculated theoretically for large light-water reactors, on the basis of nuclear physics and engineering data. According to this, an emission of about 10 Ci/GWa [1)] of C-14 is to be expected, while the C-14 component present in the nuclear fuel would be liberated at latest during the reprocessing of the fuel.

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1) 1 GWa = 1000 MW.a; this corresponds to the annual electrical power generated by a 1250 MW nuclear power plant with a loadfactor of 80%.

For 10 Ci/a of CO_2-bound C-14 liberated under normal propagation conditions via a stack 100 m in height, the maximal radiation exposure by ingestion is calculated to be some tenths of a millirem per annum. The radiation exposure by ingestion comes about when the liberated C-14 is incorporated through photosynthesis into vegetable matter which serves as food.

The radiological significance of the liberation of C-14 at nuclear power plants and reprocessing facilities was first fully recognized only in recent years, for example, by Magno (MAG 74) and Martin (MAR 74/1, 74/2). The calculation of the total dose commitment through the liberation of C-14 at nuclear power plants showed that C-14 is to be reckoned as one of the most important nuclides as regards the radiation exposure of the whole population. The reasons for this are:

(i) the long persistence of C-14 in the biosphere;

(ii) the fact that through photosynthesis CO_2-bound C-14 passes directly into vegetation and hence into animals which serve as food;

(iii) the fact that with present-day retention techniques in nuclear plants gaseous C-14 compounds escape almost entirely.

In December 1976 a working-party of the World Health Organization discussed an investigation by Pochin (POC 76) which shows up the significance of the collective dose commitment due to C-14 from nuclear power generation. The report of the working-party was published in 1977 (WHO 77). An Euratom report by Kelly et al. (Kel 75) published at the end of 1975 likewise discussed the regional and global radiation exposure in the European Community due to emissions of C-14 from nuclear plants. On the basis of the papers published in 1974 and 1975, Schwibach and Jacobi reported in January 1976 on the significance of the collective dose commitment due to C-14 from nuclear power plants and reprocessing facilities in comparison with the collective dose commitment due to the liberation of other long-lived radionuclides and with the natural radiation exposure (SCH 76).

The possible genetic and somatic effects of an increase in the C-14 content of the biosphere have been discussed in various publications, e.g., by Pauling (PAU 78). The U.S. Environmental Protection Agency (USEPA 76) published a report in November 1976 on the environmental and radiological protection requirements for normal operation of activities in the uranium fuel cycle. This report gives a number of possible serious health hazards due to long-lived radionuclides from the uranium fuel cycle. This estimate gives for C-14 values higher by whole orders of magnitude than for other long-lived radionuclides, because retention of the C-14 in the reprocessing facilities is not assumed in the study. In the paper by a working-party of the World Health Organization (WHO 77), on the other hand, a 90% retention of C-14 in the reprocessing facilities was assumed.

By means of the risk factors given, for example, in (IRCP 26), the number of somatic and genetic effects to be expected from the values of the radiation exposure can be specified. This new concept for assessing radiation hazards due to the formation of C-14 at nuclear installations has been used, e.g., in a study by Killough and Till (KIL 78/2).

In recent years, therefore, new measurements and investigations of the emission of C-14 at nuclear installations in various places have been conducted.

As early as March 1974 the Federal Minister of the Interior had given the Federal Health Office an instruction under the terms of the research project "Inquiries and studies into the surveillance of radioactive emissions at nuclear installations" to develop and apply methods of measuring C-14 in the vicinity of nuclear power plants. After the conclusion of the preliminary experiments the Federal Health Office was able to carry out the first test measurements at nuclear power plants in April 1976. Later in June 1976 collaboration between the laboratories of the Nuclear Research Centre, Karlsruhe, and Heidelberg University began. As a result in two conferences at the Federal Health Office in Neuherberg still in 1976, the existing

knowledge, the measuring processes to be adopted, the necessary measuring devices, and programmes for measurements were discussed. The Nuclear Research Centre, Karlsruhe, undertook the measurement of CO_2-bound C-14 at its installations, while the Institute for Environmental Physics, Heidelberg, obtained a research contract from the Federal Minister of the Interior to measure C-14 in environmental samples. The increase theoretically to be expected in the C-14 content in vegetable matter at the maximum points, an increase which amounts only to a small percentage of the present value, can be detected by measurement techniques only if all changes in C-14 due to local conditions (e.g., local depression due to fossil carbon from combustion gases) are taken into account.

The information at present available about emission measurements and the methods of measurement to be adopted are discussed in more detail below.

The results of investigations on vegetable matter from the immediate vicinity of nuclear power plants have in the meantime been published in (LEV 78).

2. Survey of the literature up to the beginning of 1976

2.1. Rates of formation and liberation of carbon-14

In nuclear reactors C-14 is formed by neutron reactions in the fuel, in the structural materials and in the cooling medium, and by ternary fission. Fig. 1 shows the nuclear reactions which contribute to the formation of C-14 (BON 73).

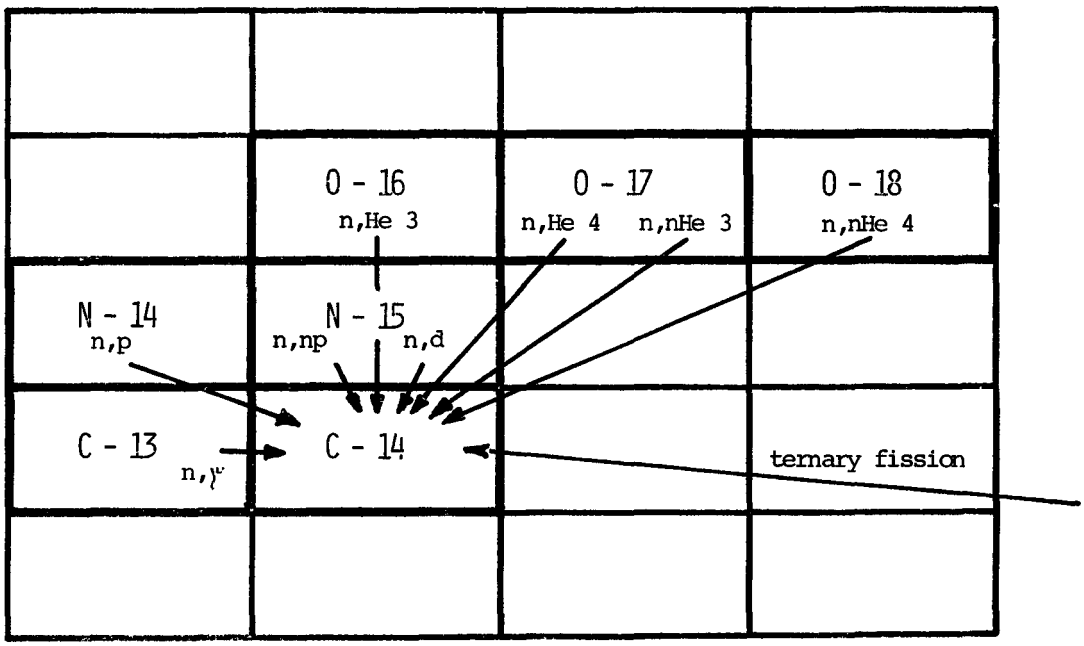

Fig. 1: Nuclear reactions which contribute to the formation of C-14

The following processes of C-14 formation are important in light-water reactors:

N-14 (n,p) C-14	effective cross-section	1,8 barn
O-17 (n,α) C-14	effective cross-section	0,24 barn
C-13 (n,γ) C-14	effective cross-section	0,0009 barn
ternary fission:	yield $1{,}7/10^6$ fissions.	

N-14 and C-13 appear in small amounts as impurities in the fuel, the structural materials, and the cooling medium.
O-17 is present naturally as 0,039% of the oxygen in the fuel

(UO_2) and in the cooling medium (H_2O). (The formation of C-14 from N-14 in the air outside the pressure vessel is negligibly small compared with its formation inside the reactor core.)

The rate of C-14 production through neutron processes is calculated by the well-known activation equation

$$R = N.\sigma_{eff}.\phi_{th}$$

where N is the number of target atoms available for activation, σ_{eff} is the effective cross-section, and ϕ_{th} is the mean flux of thermal neutrons in the active core volume.

The contributions of the separate mechanisms of production to the rate of C-14 formation depend on various factors. In particular, the nitrogen-14 component and the amount of cooling medium present in the active core have a large effect on the rate of formation.

Calculations of the rates of formation and liberation of C-14 were carried out in 1973 for high-temperature reactors (BON 73) and in 1974 for light-water reactors (BON 74). In a Russian article (RUB 73) of 1973 emission rates of about 300 Ci/GWa were calculated from measurements for light-water reactors. (This paper will not be discussed in more detail here, because the particulars regarding the sampling and measurements are not given.) In an estimate by Magno (MAG 74) an overall formation rate of 50 Ci per GWa is given for reactors without specifying the details of the calculation, while in the papers by Martin (MAR 74/1, MAR 74/2) a mean rate of formation is established as 30 Ci/GWa.

More recent calculations of the rate of C-14 formation in light-water reactors were carried out by Hayes (HAY 75), by Fowler (FOW 76) and by Davis (DAV 77).

In the papers mentioned the formation of C-14 in the cooling medium and in the fuel are calculated for the separate processes. The rates of formation of C-14 given in these papers agree to within a factor of 2. It turns out that, in the fuel, the N-14 impurities make the greatest contribution to the rate of

formation of C-14, while in the cooling medium the reaction with O-17 makes the greatest contribution.

A synopsis of the published rates of C-14 formation is given in Table 1. The formation rate in the cooling medium, given in the papers by Hayes, Fowler and Davis, lower by a factor of 3 for pressurized-water reactors than for boiling-water reactors, is to be attributed to the fact that, in boiling-water reactors, a greater volume of cooling medium is available in the active core for the formation of C-14 through the (n,α) reaction with O-17 than in pressurized-water reactors.

In the 1975 paper by Kelly et al. (KEL 75) the rate of formation of the C-14 produced in the cooling medium of nuclear reactors was derived from the measurements made by Kunz (KUN 74) (16 Ci/GWa for BWRs, 6 Ci/GWa for PWRs), while the rate of C-14 formation in the fuel was found by calculation. Similarly, in the 1977 report of the Scientific Committee of the United Nations on the effects of ionizing radiation, the rates of C-14 formation are given separately for PWRs and BWRs (UNSCEAR 77).

The above-mentioned papers on the rate of C-14 formation in light-water reactors agree in finding that while the C-14 formed in the cooling medium is liberated during the reactor's operation, the C-14 activity formed in the fuel has to be reckoned with only when the fuel elements are dissolved for reprocessing. The liberation of the C-14 takes place almost exclusively via the effluent gas, both at the reactor and at the reprocessing installation. Liberation via liquid effluent is, by contrast, negligibly small. More details about this are given in Section 2.4.

The rates so far published of formation and liberation of C-14 at reactors and reprocessing plants are grouped separately in Table 1 for boiling-water reactors and for pressurized-water reactors. The reported data were converted to the values corresponding to the generation of 1 GWa of electricity. (1 GW thermal output of a reactor was converted to GW output of electricity by assuming a thermal efficiency of 33%.)

Table 1: Synopsis of calculated rates of formation and liberation of C-14 at light-water reactors (in Ci per GWa of electricity generated)

I. Boiling-water reactors

Author	Cooling medium	Fuel	Total rate of Formation
	Liberation from reactor	Liberation at reprocessing plant[1)]	
	Ci/GWa	Ci/GWa	Ci/GWa
Bonka 1974	11,2	18,9	30,1
Hayes 1975	11,5	32,7	44,2
Fowler 1976	9,1	22	31,2
Davis 1977	4,7	17,6	22,3
Kelly 1975	16	13,6	29,6
UNSCEAR 1977	8	16	24

II. Pressurized-water reactors

Author	Cooling medium	Fuel	Total rate of Formation
	Liberation from reactor	Liberation at reprocessing plant[1)]	
	Ci/GWa	Ci/GWa	Ci/GWa
Bonka 1974	11,1	16,1	27,2
Hayes 1975	3,4	11,6	15,0
Fowler 1976	3,3	22	25,3
Davis 1977	5	18,8	23,8
Kelly 1975	6	13,6	19,6
UNSCEAR 1977	5	12	17

1) Assuming no C-14 retention.

2.2. Liberation of C-14 in reprocessing installations for spent nuclear fuel

The first detailed prediction of C-14 release in the dissolution of spent nuclear fuel in a reprocessing plant were given by Fowler (FOW 76) for the plant at Barnwell, USA. It is assumed in this report that the C-14 present in the fuel is liberated as CO_2 in the dissolution process; the estimated amount is 1000 Ci/a. In the EPA study of 1976 (USEPA 76) the question was already raised whether the resulting C-14 should not be retained.

A report in 1976 from Oak Ridge (KIL 76) discussed the environmental effects of the liberation of C-14 at a nuclear fuel reprocessing plant if the fuel comes from a high-temperature reactor. Here a liberation of 5000 Ci/a was assumed.

Another study from ORNL (TEN 76) of the radiological effects of reprocessing nuclear fuel for sodium-cooled fast-breeder reactors considered the adoption of plutonium uranium carbide and plutonium uranium nitride nuclear fuels to enrich mixed-oxide nuclear fuels. The annual output for a plant handling spent fuel equivalent to 50 GWa per year was given as follows:

- for uranium-plutonium mixed-oxides fuel at 20 ppm N_2-content --- 241 Ci;
- for uranium-plutonium carbide fuel at 0 ppm N_2 --- 1 Ci, and at 150 ppm N_2 --- 2020 Ci.

- Very high values are calculated for uranium and plutonium nitride fuels, viz., 82 700 Ci for nitrides having the natural mixture of nitrogen isotopes, 17 000 Ci for an enrichment in N-15 to 99%, and 8 780 Ci for 100% N-15. Here again a retention factor of 10^2 by separation of the CO_2-bound C-14 as calcium carbonate was assumed to be attainable. This may be compared with a calculated value for C-14 of 241 Ci for mixed-oxide fuel with 20 ppm nitrogen content.

A further study from ORNL (TEN 78) was concerned with the

radiological effects of using thorium and uranium fuel in the core and thorium carbide in the blanket of a fast-breeder reactor. In the reprocessing of the nuclear fuel a liberation of large amounts of C-14 in the gaseous effluent was expected. The adoption of an absorption process using carbon fluoride compounds should remove 99,99% of the CO_2-bound C-14 from the gas. The CO_2 would be separated and stored as calcium and barium carbonates. However, for the purpose of the study a retention factor of only 10^2 instead of 10^4 was adopted. An output of 3 750 Ci per 50 GWa generated electricity was estimated for the reprocessing plant. The rate of liberation would then amount to 37 Ci/a.

2.3. Measurements of the emission of C-14 in effluent gas from light-water reactors

2.3.1. American investigations

(i) The Yankee Rowe study

After the previous fundamental investigation into surveillance of the emission of radioactive material from nuclear power stations carried out at the Dresden plant (KAH 70), the U.S. Federal Health Service (Kahn and coworkers) carried out a further study (KAH 71) at the Yankee Row nuclear power plant, a pressurized-water reactor of 185 MWe output.

In June 1969 gas samples were taken from the off-gas tanks. During the determination of Kr-85 by means of a liquid scintillation counter, Moghissi found C-14 in the gaseous state in the samples. The activity concentration of C-14 amounted to about 8.10^{-4} Ci/m^3. Further measurements of off-gas samples from the primary coolant circuit under pressure loading showed about 10^{-3} Ci/m^3 of gaseous C-14 activity. Similarly, a C-14 activity concentration of about 10^{-6} and 10^{-9} was measured in the containment atmosphere.

In the report an emission of altogether about 0,3 Ci/a gaseous

C-14 compounds was calculated, of which 0,11 Ci/a should come from the off-gas tanks and 0,18 Ci/a from the containment atmosphere during ventilation before changing the fuel elements.

It has become known that a second path for liberation of C-14 in the effluent gas is the off-gas exhaust from the secondary side condenser in the turbine hall. During leakage into the secondary side this emission takes place continuously. It was found, however, that exhaust gas samples from the stack show in particular cases a 12 times greater C-14 activity concentration than the values calculated from individual measurements at the secondary side condenser.

(ii) The Haddam Neck study

Another investigation of this kind was carried out by the U.S. Federal Health Service at the Haddam Neck nuclear power plant, a pressurized-water reactor of 590 MWe (KAH 74). The measurement were made for samples from the off-gas tanks, the containment atmosphere, the off-gas exhaust from the secondary side condenser and the air from the stack (from the reactor building) from the auxiliary plant building and from the turbine hall. The activity concentration in the containment atmosphere, for example, amounted to 2.10^{-6} Ci/m^3 for C-14 in the form of hydrocarbons, and 8.10^{-8} for C-14 in the form of carbon dioxide. From these measurements an emission of 0,18 Ci/a of C-14 due to ventilation of the containment atmosphere was calculated. The exhaust air from the fuel element building contributed a relatively high amount, 0,3 Ci/a of C-14. Altogether, it was calculated from these measurements that about 0,54 Ci/a of C-14 escapes through the reactor building stack.

The measurements of the activity concentration of C-14 in the stack gas from the reactor building gave values of, e.g., 3.10^{-9} , $1,7.10^{-8}$, 3.10^{-8} and 7.10^{-8} Ci/m^3. In the report it is stated that, on the basis of these individual measurements, the yearly emission of C-14 from the reactor building stack is estimated to be about 3 Ci. Other measurements are mentioned in

the report which indicate an additional emission of C-14 via the exhaust air from the auxiliary plant building and the turbine hall (the ventilation of these buildings does not take place through the reactor building stack).

A measurement in the exhaust air from the auxiliary plant building gave a C-14 activity concentration of 6.10^{-9} Ci/m^3 and in the exhaust air from the turbine hall 8.10^{-9} Ci/m^3. Hence a further emission of < 2 and < 10 Ci/a of C-14, resp. was calculated.

(iii) The Oyster Creek study

The latest published study of this sort was carried out by the Federal Health Service at the Oyster Creek nuclear power plant, a boiling-water reactor of 640 MWe output (BLA 76).

Measurements in the reactor building, the turbine hall, and in the adjacent buildings gave for C-14 in the form of CO_2 the respective values 0,3 Ci/a, 0,85 Ci/a, and 0,03 Ci/a, and for C-14 in the form of hydrocarbons and carbon monoxide 0,05 Ci/a, 0,06 Ci/a, and 0,01 Ci/a. The total emission was therefore estimated at 3,2 Ci/a of C-14 in CO_2-form and 1 Ci/a of C-14 in the form of hydrocarbons and carbon monoxide.

However, separate measurements in the stack of the nuclear works lead to the estimate for the yearly emission of 8 Ci of C-14 in CO_2-form and 1 Ci of C-14 in the form of hydrocarbons and carbon monoxide.

(iv) Investigations at the nuclear power plants at Ginna, Indian Point, and Nine Mile Point

Investigations over several years into the occurrence of C-14 in gaseous effluent from various pressurized-water reactors and boiling-water reactors were carried out by the radiological protection laboratories of the Health Authorities of New York State (Matuszek et al.). In the first report (MAT 73) the estimated emission of C-14 in the form of methane and carbon

dioxide, based on the analysis of various samples, was given. With PWRs most of the C-14 was in the form of methane, with BWRs most was in the form of carbon dioxide. In this report the following values are given:

BWRs	(910 MWe)	0,4 Ci/a
PWR	(805 MWe)	> 2 Ci/a.

It is also pointed out that the values for the CO_2 form are probably too low because there was contact with water when the samples were taken.

The second report (KUN 74) deals with the measured results for samples from the off-gas tanks and the containment atmosphere at the pressurized-water reactors at Ginna (450 MWe) and Indian Point, 1 and 2. More than 95% of the C-14 activity lay in the hydrocarbon fraction. The concentration in the off-gas tanks was 2.10^{-4} to 9.10^{-4} Ci/m^3. The estimated emission values[1] per month from the off-gas tanks amounted to:

0,064 Ci	in February	1973
0,23 Ci	in October	1973
0,11 Ci	in February	1974
0,12 Ci	in June	1974.

The emission from Ginna calculated for these values was 1,5 Ci/a.

In the containment atmosphere an activity concentration of 1.10^{-6} Ci/m^3 was measured; this implies an additional emission of 0,28 Ci/a. The measured values of C-14 from the off-gas tanks at Indian Point were similar. However, the measurements in the ventilation from the buildings and in the stack were not reported.

The third report dealt with measurements at the boiling-water reactor at Nine Mile Point (610 MWe) (KUN 75). The samples were taken from the off-gas condenser; 95% of the C-14 was in the form of CO_2. The emission according to the measured values amounted to 8 Ci/a. Measurements in the air ventilated from the buildings and in the stack itself were not reported.

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1) Although an averaging effect was inherent in the sampling from the off-gas tanks appreciable scatter can be observed.

(v) Annual emission at Yankee Rowe

For the nuclear power plant at Yankee Rowe data for the annual emission of C-14 are also available in the AEC and NRC reports. According to these the amounts of C-14 emitted were:

in 1972	1	Ci	(AEC 73)
in 1974	0,5	Ci	(NUR 76)
in 1975	1,6	Ci	(NUR 77), and
in 1976	0,6	Ci	(NUR 78).

Further details about the sampling and the measurements are not given.

(vi) Summary of the American investigations

The measurements in American BWRs and PWRs reported above in (i) to (v) are collected together in Table 2. This gives the reported annual emissions of C-14, based in part on estimates.

For comparison purposes the emission per GWa of electricity generated has also been calculated from the installed electrical output. These correspond to the annual emission from a nuclear power plant of 1 250 MW electrical output working with a load factor of 80%. For the Yankee Rowe station a footnote to the table gives the annual emission in Ci/GWa calculated for the actual power generated.

It seems at present to be more accurate to relate the C-14 emission to the actual power generated during the year. However, the errors would increase, if the reference period were shorter than a year, especially if there is a time lag between formation and liberation, for example, if further C-14 is emitted while a plant is closed down. Exact values are obtained only by comparing the discharge and the power generated over long periods, going back if possible to the start-up date.

Table 2. Synopsis of the emission of C-14 from American nuclear power plants

Site/sampling location	Reported annual emission Ci	Calculated emission per GWa (installed capacity) Ci/GWa
I. Boiling-water reactors		
1. Oyster Creek (640 MW)		
a) Gaseous effluent plant	3)	
Reactor building	0,36)	8
Turbine hall	0,9)	
Adjacent buildings	0,04)	
b) Stack	9	18
2. Nine Mile Point (610 MW)		
a) Off-gas installation	8	16 [1]
b) Stack	No data	
1) This value is also given in (KUN 75).		
II. Pressurized-water reactors		
1. Yankee Rowe (185 MW)		
a) Off-gas tank	0,11)	
Containment	0,18)	2
b) Annual emission 1972	1	7 [1]
1973	0,5	3 [1]
1975	1,6	11 [1]
1976	0,1	0,7 [1]
2. Haddam Neck (390 MW)		
a) Off-gas tank	0,08)	
Containment	0,18)	1
Fuel element building	0,3	
b) Reactor building stack	< 3	< 6
c) Auxiliary plant buildings	< 2	
Turbine hall	< 10	
3. Ginna (450 MW)		
a) Off-gas tank	1,5)	
Containment	0,28)	5 [2]

1) Taking account of the actual generated power, as given in (AEC 73, NUR 76, NUR 77, NUR 78) 13 Ci/GWa (1972), 5 Ci/GWa (1974), 13 Ci/GWa (1975), 1 Ci/GWa (1976)

2) In (KUN 74) 6 Ci/GWa is given.

2.3.2. European investigations

(i) Investigations at the nuclear research centre, Karlsruhe (SCH 77/1)

Measurements of CO_2-bound C-14 were made at the nuclear power plant Obrigheim (pressurized-water reactor, 345 MWe) over a 6 week period from Sept. the 3oth, to Nov. the 12th, 1976. The values lie in the range 0,4 to $1,8.10^{-9}$ Ci/m^3 with a mean value of $1,04.10^{-9}$ Ci/m^3. From this the annual emission of CO_2-bound C-14 at the Obrigheim nuclear power plant was estimated as 1,1 Ci/a. Simultaneous measurements in the ventilation air from the equipment rooms showed that the contribution from this part of the facility was negligibly small at 0,025 Ci/a.

Further measurements were carried out at the nuclear power plant Biblis Block A (pressurized-water reactor 1204 MWe). Concentrations of CO_2-bound C-14 of between 0,2 and $1,1.10^{-9}$ Ci/m^3 in the stack effluent gas were measured between Oct. the 8th, and Nov. the 11th, 1976. From the mean concentration over this period, $4,4.10^{-10}$ Ci/m^3, an annual emission of 0,7 Ci/a of CO_2-bound C-14 at the Biblis Block A plant was estimated.

It is to be noted that these measurements embrace only the CO_2-bound emission, not the total emission of C-14. Normalized to a generated output of 1 GWa, the following annual emissions result from the discharges estimated by Schüttelkopf:
Obrigheim: 3,2 Ci CO_2-bound C-14 per GWa
Biblis A: 0,6 Ci CO_2-bound C-14 per GWa.

(ii) Investigations in Switzerland

In the annual report of the Swiss Commission for surveillance of radioactivity for the year 1977 the results of random sampling of effluent gas are given. The resulting estimates of the annual discharge from the Mühlberg nuclear plant are of the order 4 Ci C-14. The question whether a small increase in the C-14 content of samples of plants and foliage can be detected is also discussed (KUE 77).

2.4. Discharge of C-14 with liquid effluent

Reports on the discharge of C-14 compounds in the liquid effluent from nuclear power plants are given in several publications. Thus, in the EPA study (FOW 76), a release of about 0,05 Ci/a is calculated for boiling-water reactors; that is less than 1% of the corresponding emission in the effluent gas. For pressurized-water reactors the value is still lower, about 0,03 Ci/a. Matuszek carried out some experimental work on this subject. The C-14/CO_2 levels in steam generator blowdown liquors of boiling-water and pressurized-water reactors lay in the range of 10^{-7} to 10^{-5} Ci/m^3. Similar values were also established in samples from the filters of the spent water reprocessing system and in ion exchangers. Values of the C-14 discharge from the Yankee Rowe plant are also contained in the NRC annual report; according to this the discharge of C-14 from the Yankee Rowe plant was:

3,1 mCi in 1974 (NUR 76)
3,8 mCi in 1975 (NUR 77)
6,1 mCi in 1976 (NUR 78).

Investigations by the Institute for Water, Soil, and Air Hygiene, of the Federal Health Office, gave the following annual discharges in liquid effluent in 1977 for example (GAN 78):

Nuclear power plant with pressurized-water reactors:

Stade	6,6 mCi of which	0,1 m Ci is organically bound
Biblis A	1,4	0,1
Neckarwestheim	1,4	0,1

Nuclear plant with boiling-water reactors

Brunsbüttel 39 mCi of which 0,02 mCi is organically bound.

Further investigations at the nuclear power plants Gundremmingen, Obrigheim, and Würgassen over separate periods of a few months gave relatively pronounced variations of the C-14 concentration in the effluent.

2.5. Measurements at the reprocessing plant, Karlsruhe

Measurements of the emission of C-14 by reprocessing plants have been made in respect of that at Karlsruhe.

From July 1976 to June 1977 Schüttelkopf made C-14/CO_2 measurements in gaseous effluent from the plant at Karlsruhe; the results are reported in the papers (SCH 77/1), (SCH 77/2), (SCH 77/3). During this period, fuel from a pressurized-water reactor (the Obrigheim plant) with a burnup of 24,5 GWd/t and from a boiling-water reactor (the Gundremmingen plant) with a burnup of 19,2 GWd/t was processed.

From the measured mean rates of C-14 emission, viz.

33 mCi/dissolution for the dissolution of fuel from the pressurized-water reactor

and 39 mCi/dissolution for the dissolution of fuel from the boiling-water reactor

Schüttelkopf obtained, by extrapolating from the charge burnup and the amount of fuel dissolved, and by using the ratio 0,33 GW_e/GW_{th}, the rate of emission for 1 GWa of power generated.

From 5 separate measurements during 10 dissolutions of fuel from a pressurized-water reactor a mean value of

$$12,4 \pm 0,3 \text{ Ci/GWa}$$

was calculated, and from 17 separate measurements during 57 dissolutions of fuel from a boiling-water reactor a mean value of

$$13,7 \pm 0,7 \text{ Ci/GWa}$$

was obtained.

3. Description of the methods of sampling and measurement for C-14 determinations

3.1. General survey

Carbon-14 emits only beta-rays, with a maximum energy of 158 keV. With the measuring instruments at present in use for monitoring gaseous effluents --- these are, in general, beta integrating instruments --- it is not possible to detect C-14, or at least, to distinguish C-14, because there is a significantly greater noble-gas activity. To detect C-14 special methods of sampling and measurement are necessary. They depend in principle on a specific separation of the C-14 compounds, followed by beta measurement.

Measuring methods, particulary for very low C-14 concentrations, have been known for a long time from C-14 dating techniques and studies of the atmospheric distribution of C-14 (LIB 55, MÜN 63/1, MÜN 63/2). In these C-14 is measured as a gas (either extremely pure CO_2 or CH_4) in beta-sensitive gas proportional counters. The counter tubes, some of which can be operated at above the atmospheric pressure, can take several litres of the gas, corresponding to about 5 g of carbon.

The samples of CO_2 gas are taken by freezing or by absorption in caustic soda solution (POV 68) or by molecular sieves (ASH 72). The carbon dioxide is driven off from the samples, purified, and fed into the counter. However, this technique is very expensive and time-consuming and is not suitable for routine use.

The development and improvement of the liquid scintillation measuring technique has made an essential contribution to reducing the cost of C-14 measurements and has led to methods for routine measurements, not least for radiological protection purposes.

This technique offers many possibilities as regards the physical-chemical form in which the C-14 can be sampled.

Inorganic absorbers for CO_2 (like NaOH) or organic absorbers (like phenylethylamine or ethanolamine) can be utilized directly in the measuring test (HOR 68). However, for long times of observation or bigger throughputs of air the solutions become unstable and do not give representative results.

Good reproducibility at a relatively low analytic cost is achieved by measurement of solid $BaCO_3$ (TUR 69, ANG 78, GOL 75, RUD 76).

For this purpose the CO_2 is separated in caustic soda solution, precipitated as $BaCO_3$ and finely ground, and then measured as a suspension in a liquid scintillation counter.

Other possible C-14 compounds are transformed into CO_2 by catalytic oxidation and prepared as above. This method was experimentally tested by us and optimized as regards both completeness of separation of the CO_2 and stability and reproducibility of measurements. It is described in more detail below.

3.2. Sampling the effluent gas from nuclear power plant

C-14 measurements in gaseous effluent of light-water cooled reactors have been carried out since April 1976 by the Federal Health Office in collaboration with the operators. In these measurements a method was used at first which allowed the C-14 to be measured both in the form of CO_2 and in forms such as CO, CH_4, etc. For this purpose samples of about 800 l of the effluent gas were forced into pressure bottles by H.P. compressors in the nuclear plants, and the separation of the C-14 compounds was carried out in the laboratory.

It was found that the use of the H.P. compressor for taking samples of the gas could produce a contamination from the preceding samples (a memory effect). For this reason, from 1977 onwards the method of taking samples of the effluent gas has been changed and now gas-tight containers of 40 l capacity

("plastigas" test gas containers made by the Linde firm) are used; these are filled with the effluent gas by means of a membrane pump delivering 1 m^3/h. It has been possible to establish by a large number of tests that, using this method of sampling, no contamination of the apparatus for taking the samples occurs. Before a sample is taken, some methane carrier-gas is put into the empty containers, in order to be able to check the yield from the catalytic combustion of the non-CO_2-bound C-14 compounds. A carrier-gas is not needed for the CO_2-bound C-14, because there is sufficient CO_2 present in the effluent gas. It takes some 10 to 20 minutes to fill each container with sample gas. The equipment used in this method of taking samples is shown in Fig. 2.

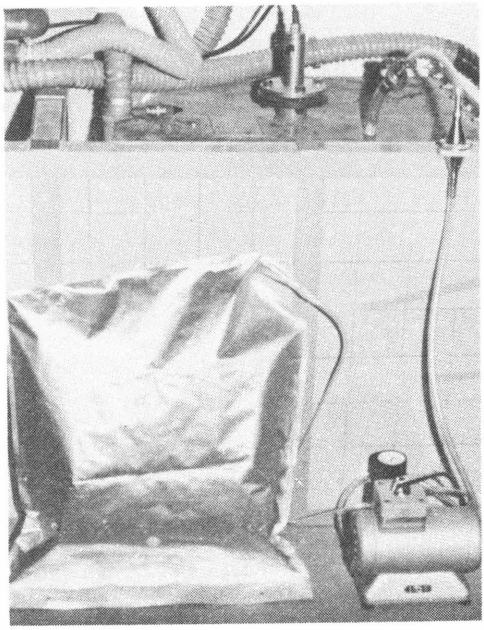

Fig. 2. Taking samples of the effluent gas in 40 l bags

3.3. Separation of the chemical forms of C-14 in effluent gas samples

The gas samples taken at the nuclear power plants were pumped through the C-14 analysis apparatus in the laboratory at a rate of about 0,5 l/min.

After fine filtering through an aerosol filter the CO_2 was removed from the sample by passing the gas through wash bottles with frits, and then the CO and hydrocarbons (in CH_4 as carrier) were oxidized catalytically (catalyzer/furnace at 800°C) and the CO_2 formed was separated in further 2 wash bottles. The apparatus is shown diagrammatically in Fig. 3.

3.4. Continuous sampling of CO_2-bound C-14

Because the liberation of C-14 in the effluent gas is not continuous, the measurements by the methods described in Section 3.2. show marked scatter of the C-14 concentrations. We therefore tried to set up a direct method of sampling CO_2 from the effluent gas, which would make it possible to sample over a long period or in continuous operation. The sampling method depends on the separation of CO_2 from the effluent in NaOH bubblers the CO_2 being separated according to the following reaction:

$$2\ NaOH + CO_2 \longrightarrow Na_2CO_3 + H_2O.$$

For direct sampling a complete separation of the CO_2 from a bleed flow of the effluent is to be sought. There are about 330 ppm of CO_2 present in the effluent (as in the surrounding environmental atmosphere); hence it has been found that the following conditions are optimal:

air throughout 1 - 2 m^3
wash bottles with frits or Friedrichs-type bottles
absorbent solution 1 mol NaOH.
An air throughput of 1 m^3 yields about 3 g of barium carbonate if separation is complete.

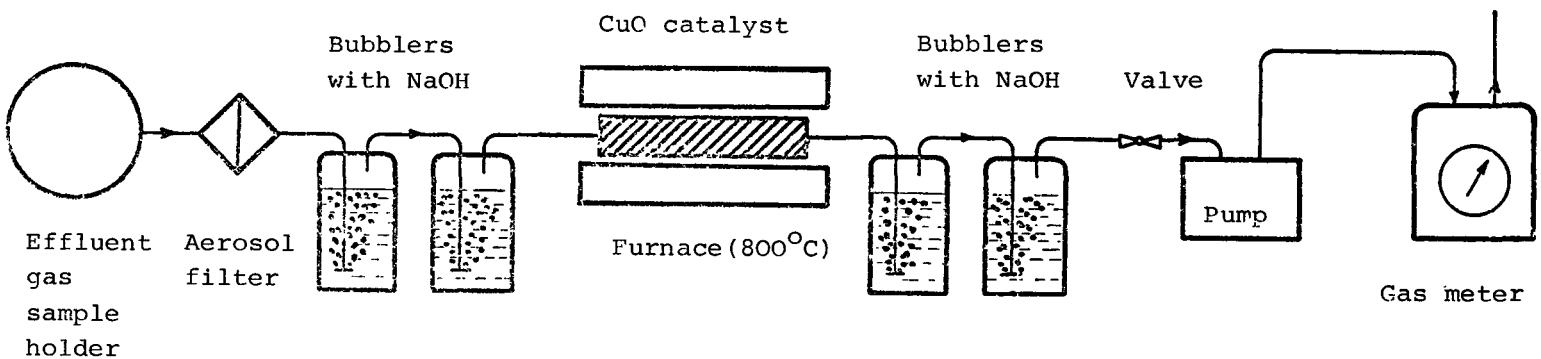

Fig. 3 Schematic diagram of the apparatus for separating the CO_2, CO and hydrocarbons from the effluent gas sample

The amount and molar strength of the NaOH solution were chosen so that complete separation is assured even towards the end of sampling (a 4 times excess of caustic).

Whenn 100 ml of 1m NaOH are used, the throughput of gas should be maintained at about the following rates, depending on the duration of the sample:

daily sample	about 30 l/h
weekly sample	about 5 l/h
monthly sample	about 1 l/h

In designing the sampling apparatus it is necessary that one ensures that the gas throughput has sufficient opportunity of reacting with the caustic solution. This can be achieved either by breaking up the gas flow into fine bubbles by means of gas-washing bottles with fine-pore frits (G1) or by keeping the gas stream in the caustic solution longer by using Friedrichs-type gas-washing bottles.

In our experience the latter, in which the gas bubbles are led in a spiral through the caustic solution, are better for throughputs of less than 100 ml/min, but gas-washing bottles with frits are better for throughputs above 100 ml/min. In both arrangements 97% separation of the CO_2 has been achieved.

Fig. 4 is a schematic diagram of the apparatus for weekly and monthly total sampling.

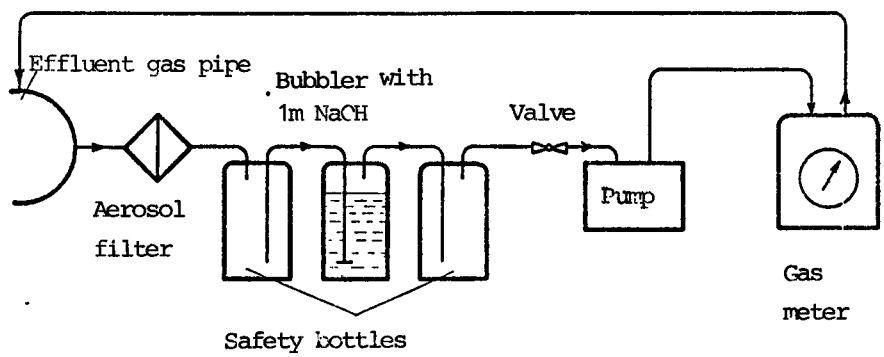

Fig. 4. Schematic diagram of the apparatus for sampling CO_2 in the effluent gas from nuclear plant

3.5. Continuous sampling of C-14 compounds in all forms

In accordance with the principle of the apparatus for analysis described in section 3.2., nowadays complete on-line separation of all forms of C-14 compounds are carried out at the nuclear power plants at Obrigheim and Stade, where there are pressurized-water reactors. No carrier gas, however, is added to the effluent gas. At Stade, part of the effluent gas is led off at the rate of about 0,5 l/h and monthly samples are obtained; at the Obrigheim plant a throughput of about 5 l/h for weekly sampling is installed.

Up to now, no difficulties have occurred apart from irregularities of the throughput of effluent. Comparative measurements of random samples at the Obrigheim plant have shown that, at this low rate of throughput, the catalytic combustion of the CO and hydrocarbons takes place completely.

3.6. Chemical separation for measurement of C-14

After the passage of the gaseous effluent sample the C-14 is present in the NaOH absorber solution as dissolved NA_2CO_3. By adding 0,5 m $BaCl_2$ solution $BaCO_3$ is pre cipitated. The precipitate is filtered off with a fine-pore frit filter, dried until its weight is constant, and the yield is then determined. A maximum of 2 g of the previously fine-ground barium carbonate is weighed into a 20 ml counting vial, mixed with 20 ml of scintillator solution (Instagel), and after ultrasonic treatment is measured in a liquid scintillation counter.
Fig. 5 shows the equipment used in preparing the sample.

The advantage of this measuring technique lies, on the one hand, in the relatively simple preparation of the sample, and, on the other hand, in the high counting efficiency (55 to 65 percent, depending on the $BaCO_3$ weight).

Another method for determining C-14 as $BaCO_3$ is given in the report (SCH 77/2) from the Nuclear Research Centre, Karlsruhe.

In this the measurement is made on a pressed tablet of $BaCO_3$ as an "infinite depth" beta source in a low-level beta counter. A counter efficiency of about 2% for the C-14 is achieved in this method.

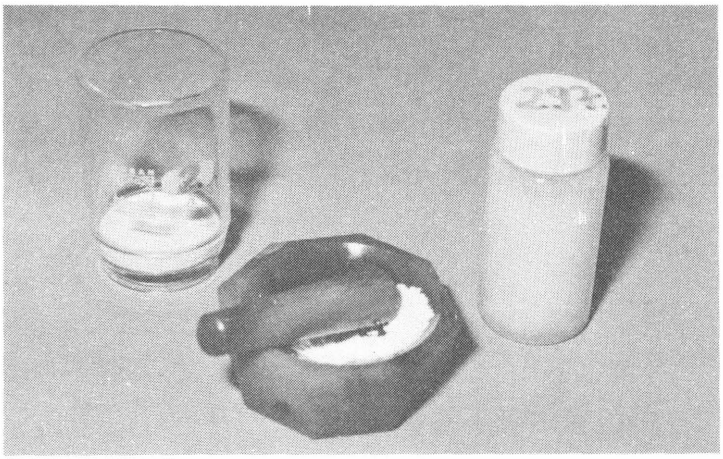

Fig. 5 Preparation of a C-14 sample: glass frit with $BaCO_3$ precipitate; agate mortar with finely ground $BaCO_3$; preparation ready for measurement.

3.7. Evaluation of the measurements

The activity concentration of the C-14 in the effluent gas is calculated from the relation

$$K = \frac{R_N \cdot 10^{-12}}{n_{ch} . n_Z . V . 2,22}$$

where K is the C-14 concentration in the effluent gas, in Ci/m^3,

R_N is the net pulse rate in cpm,

n_{ch} is the chemical yield,

n_Z is the efficiency of the counter,

and V is the volume of the gas sample.

The limit of detection by this method is about $20 pCi/m^3$ with an initial volume of 80 l (2 containers of test gas), or about

2 pCi/m^3 with an initial volume of 1 m^3 (e.g., in direct separation in NaOH).

The mean error of a measured value of the C-14 concentration in the effluent gas is made up of the statistical counting error and the errors which arise in the determination of the count, in the preparation of the sample for measurement, and in the measurement of the volume of the gas.

In the activity range from 10^{-9} to 10^{-10} Ci/m^3 the statistical counting error is negligible for measurements lasting more than 100 minutes, while near the lower limit of detection this error predominates. The non-statistical errors are estimated at $\pm$ 10%.

To check the measured results some of the following additional measurements should be carried out on samples selected at random:

1. Gamma spectrometry of the sample on a Ge (Li) detector to check whether gamma radiation may be present;
2. Measurement of a beta spectrum with a liquid scintillation counter either
   a) by attaching a multi-channel analyzer, or
   b) by stepwise displacement of the counter window on the liquid scintillation counter and comparison with a standard sample;
3. Repeated measurement of the sample after a longer time interval. The samples are stable for months. A reduced count rate in the later measurement would indicate short half-life nuclides were originally present.
4. Additional check measurements can be carried out on the remaining amount of $BaCO_3$ not initially used, e.g., by dissolving the $BaCO_3$, reprecipitating, and final measurement.

4. Results of measurements by the Federal Health Office

4.1. General

In this section we have collected together the results of some 700 individual measurements of the C-14 concentration in the effluent gas from the nuclear power plants Gundremmingen, Lingen, Würgassen, Brunsbüttel, Obrigheim, Stade, Biblis A, Biblis B, and Neckarwestheim.

The results of the first measurements from the year 1976 were published in the report "Investigations of the emission of carbon-14 in the effluent gas from nuclear power plants" (RIE 76) in December 1976. From 1977 augmented methods of sampling with continuous separation of CO_2-bound C-14 were initiated, because it had been found that it is difficult to estimate the annual emission of C-14 in the effluent gas from a few individual measurements. The measured results accumulated in the Federal Health Office up to August 1977 were published in a second report (RIE 77).

In this paper we refer to all these results and report on the further measurements carried out by the Federal Health Office up to October 1978 inclusive. Investigations into the effluent gas from the reprocessing plant at Karlsruhe and into the effluent gas from a boiling-water reactor (the Brunsbüttel nuclear power plant) and from a pressurized-water reactor (the nuclear plant at Neckarwestheim) are also included.

Some essential information about the nuclear power plants included in the investigation is given in Table 3.

Regarding the results of measurements it is to be noted that measured values based on short-time sampling show marked scatter. This is due, on the one hand, to variations in the C-14 concentration due to the operating regime of a plant, and, on the other hand, to variations due to discontinuous emission even during a constant operating regime. For this reason we have tried to obtain a representive survey of the C-14 emission

Table 3: Data for the nuclear power plants

Nuclear power plant	Short title	Capacity MWe	Commissioned
I. Boiling-water reactors			
Kahl	VAK	15	1961
Gundremmingen	KRB	252	1966
Lingen	KWL	252 [1]	1968
Würgassen	KWW	670 [2]	1972
Brunsbüttel	KKB	806	1976
II. Pressurized-water reactors			
Obrigheim	KWO	345	1968
Stade	KKS	622	1972
Biblis A	KBA	1204	1974
Biblis B	KBB	1300	1976
Neckarwestheim	GKN	855	1976

1) Reactor output about 165 MWe plus fossilized-fuel superheater

2) Was operated with only 80% of the installed capacity (536 MWe)

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either by a multiplicity of separate measurements or by integrative sampling over longer periods. Table 4 gives a summary of the sampling methods carried out with the stack effluent.

4.2. Measurements of the C-14 emission at boiling-water reactors

4.2.1. Measurements of the various forms of C-14 compounds in the effluent gas

At the boiling-water reactors Kahl, Gundremmingen, Lingen, and Würgassen measurements were carried out in 1976 and 1977 of the CO_2-bound, CO-bound, and hydrocarbon-bound components of the C-14. 95% of the C-14 activity is found to be in the CO_2 form in the effluent gas.

The measured CO_2-bound fractions of the C-14 in the effluent

Table 4: Summary of the sampling methods used for the stack effluent

Nuclear power plant	Type of sampling	Frequency
Gundremmingen	CO_2 separation in NaOH; discontinuous	Once a week, over 24 hours
Lingen	CO_2 separation in NaOH; discontinuous up to September 1977	Once a fortnight, over 24 hours
Würgassen	CO_2 separation in NaOH; continuous	Monthly total samples
Brunsbüttel	CO_2 separation in NaOH; continuous	Weekly total samples
Obrigheim	CO_2,CH_4 random samples; discontinuous	1 sample per month
Stade	CO_2 separation in NaOH; continuous	Monthly total samples
	CO_2,CH_4 separation; continuous	Monthly total samples
Biblis A	CO_2,CH_4 random samples; discontinuous	1 sample per month
Biblis B	CO_2, separation in NaOH; continuous	Monthly total samples
	CO_2,CH_4 random samples; discontinuous	1 sample per month
Neckarwestheim	CO_2 separation in NaOH; continuous	Weekly total samples
	CO_2,CH_4 random samples; discontinuous	1 sample per month

gas from the individual reactors are collected together in Table 5.

Table 5. CO_2-bound fraction of the C-14 in the total emission from boiling-water reactors

Experimental atomic power plant Kahl:	98%	CO_2-bound C-14
Nuclear power plant Gundremmingen	96%	
Lingen	96%	
Würgassen	98%	

From this experience, it was decided to make only measurements of the CO_2-bound components of the C-14 in 1977 and 1978 at boiling-water reactors, since this component accounts for more than 95% of the total emission.

4.2.2. Measurements of the C-14 concentration in the effluent gas

(i) The nuclear power plant at Gundremmingen

In the effluent gas from the nuclear power plant at Gundremmingen CO_2 separation has been carried out from October 1976 for periods of 24 hours once a fortnight or once a week. During normal reactor operation with full output the C-14 concentration in the effluent gas lay in the range 1.10^{-9} to 4.10^{-9} Ci/m^3, with a mean value of $1,9.10^{-9}$ Ci/m^3; this gives a mean annual emission of 3,7 Ci/a calculated from the mean C-14 concentration and the air throughput of 220 000 m^3/h.

Relative to the installed reactor power, this corresponds to a normalized annual emission of 14,7 Ci/GWa, or 15,1 Ci/GWa if the actual power generated is considered.

The nuclear power plant was closed down on January 13th, 1977. The effluent C-14 concentration has therefore diminished almost continuously. In 1977 1,0 Ci, and in 1978 0,1 Ci were still recorded.

The individual results are collected together in the Appendix in Tables A1, A2, A3 and in Fig. A1.

(ii) The nuclear power plant at Lingen

Only 3 measurements were made in the effluent gas from the nuclear power plant at Lingen during normal working. The sampling took place in November and December 1976, over 24 hours in each case. The C-14 concentration in the effluent lay between $1,1.10^{-9}$ and $1,8.10^{-9}$ Ci/m^3. The nuclear plant at Lingen was

closed down on January 5th,1977. On January 11th, 1977 an increase in the C-14 concentration in the effluent to $3,3.10^{-9}$ Ci/m^3 was measured. From the mean of these 4 values, viz., $1,9.10^{-9}$ Ci/m^3, and the air throughput of 85 000 m^3/h an annual emission of 1,4 Ci is calculated.

The nuclear power plant Lingen is operated with a fossil-fuelled superheater; about 2/3 of the installed capacity, 252 MWe, is obtained from the reactor (165 MWe).

Relative to the installed reactor power, this corresponds to a normalized annual emission of 8,5 Ci/GW, or 9,2 Ci/GWa if the actual power generated is considered.

After January 12th,1977 a steady decrease in the C-14 concentration in the effluent was observed. In September 1977 the measurements were discontinued, because the limit of detection had been reached at this time. From the measurements in 1977 an emission of 0,2 Ci for that year was calculated. The individual results are collected together in the Tables A4, A5, A6 and in Fig. A2 in the Appendix.

(iii) The nuclear power plant Würgassen

Continuous separation of the CO_2 from the effluent gas from the nuclear power plant at Würgassen was carried out from January 1977. The absorbent solutions were changed monthly and the monthly mean value of the C-14 concentration was determined.

In 1977 and 1978 the same mean concentration of CO_2 in the effluent gas was determined, viz., $1,5.10^{-9}$ Ci/m^3. With an air throughput of 470 000 m^3/h this gives an annual emission of 6,2 Ci for both 1977 and 1978. The individual monthly values of the C-14 concentration lay between $3,9.10^{-11}$ Ci/m^3 and $2,8.10^{-9}$ Ci/m^3. During constant reactor operation (from January 1977 to June 1977 and from November 1977 to April 1978) the C-14 concentration was relatively constant (9.10^{-10} to $2,8.10^{-9}$ Ci/m^3). The lowest values occurred in the months during which the plant was at a standstill because of overhaul work

(May 1978; $3{,}9.10^{-11}$), or when the plant came back into operation after the overhaul period (October 1977; $2{,}6.10^{-10}$ Ci/m^3).

In standardizing the annual emission relative to the reactor capacity, it was taken into account that the nuclear power plant is operating only at 80% of the designed power of 670 MWe, i.e. at 536 MWe. For 1977 and 1978 a normalized annual emission of 11,6 Ci/GWa was calculated for each year. Taking the actual power generated in 1977 into account, the normalized annual emission becomes 14,3 Ci/GWa.

Data on the power generated in 1978 are at present only available for the first half year. From the emission of 3,3 Ci in this period and the actual power generated $1{,}2.10^3$ GWh, a normalized annual emission of 23,8 Ci/GWa is calculated. This high value is partly to be attributed to the fact that the plant was closed down on April 7th, but C-14 continued to be emitted. The individual results are collected together in Tables A7, A8, and in Fi. A3 and A4.

(iv) The nuclear power plant Brunsbüttel

In the Brunsbüttel nuclear power plant continuous CO_2 separation was carried out from July 1977 onwards. Up to October 1977 monthly total samples were taken, thereafter weekly total samples. In 1977 and also in 1978 the operating conditions varied considerably and the marked fluctuations in the monthly mean value of the C-14 concentrations, which lie in the range $5{,}8.10^{-12}$ to $7{,}9.10^{-9}$, are to be attributed to these circumstances.

The weekly total samples showed even stronger fluctuations (4.10^{-12} to $2{,}5.10^{-8}$ Ci/m^3). The lowest values occurred in November and December 1977 during longer shutdown periods, viz., $8{,}2.10^{-12}$ and $5{,}8.10^{-12}$ Ci/m^3 respectively. The mean C-14 concentration in the effluent gas amounted to $1{,}4.10^{-9}$ Ci/m^3 in 1977, and $1{,}5.10^{-9}$ Ci/m^3 in 1978. Using the air throughput of 350 000 m^3/h, these values give the annual emissions 4,3 Ci for 1977 and 4,5 Ci for 1978. Relative to the

reactor capacity of 806 MWe, the normalized annual emissions are 5,3 Ci/GWa for 1977 and 5,6 Ci/GWa for 1978. Relative to the actual power generated, the values are twice as great (10,9 Ci/GWa in 1977, and 12,9 Ci/GWa for the first half of 1978). The individual results are collected together in Tables A9, A10 and Fig. A5 and A6 in the Appendix.

4.2.3. Summary of the results of measurements at boiling-water reactors

The values determined for the annual emission of CO_2-bound C-14 in the effluent gas from the boiling-water reactors, and the yearly emission per GWa of power generated are shown in Table 6.

Table 6: Summary of the CO_2-bound C-14 emission in the effluent gas from boiling-water reactors

Nuclear power plant	Year	Annual emission	Annual emission relative to current generated
		Ci/a	Ci/GWa
Gundremmingen	1976	3,7	15,1
Lingen	1976	1,4	9,2
Würgassen	1977	6,2	14,3
	1978	6,2	(23,8) [1)]
Brunsbüttel	1977	4,3	10,9
	1978	4,5	12,9

1) See page 35.

It is to be noted here that the annual emissions were calculated on occasion from only a few individual measurements or from observations over short time intervals. Moreover, in 1978 the figures for the power generated at the individual plants were available only for the first half-year, so that in this case too the annual values had to be over-estimated. (These outputs at the separate stations were taken from the monthly issues of

the journal "Atomwirtschaft".)

The average over the individual nuclear power plants for the annual emission relative to the power generated is

14 Ci/GWa

for boiling-water reactors. This is equivalent to a mean annual emission of 10 Ci for a boiling-water reactor of 1000 MW nominal electrical output.

4.3. Measurements of the C-14 emission at pressurized-water reactors

4.3.1. Measurements of the various forms of C-14 compounds in the effluent gas

The measurement of the proportions of CO_2-bound, CO-bound, and hydrocarbon-bound C-14 in the effluent gas from pressurized-water reactors have shown that the proportion of each of the various forms of compounds in the total C-14 emission is subject to strong fluctuations.

From measurements of random samples the proportion of CO_2-bound C-14 in the total C-14 emission was found to vary between 0,8% and 81%. Longer methods of integrative sampling have shown a similar effect.

We have observed that if an increased rate of C-14 emission above the mean value is observed, this is always to be attributed to a greater proportion of CO-bound and hydrocarbon-bound C-14, while the emission rate of CO_2-bound C-14 remains comparatively constant.

Measurements of samples from the off-gas plant show, in agreement, that in this part of the flow only about 0,7% of C-14 is CO_2-bound. Since with pressurized-water reactors the emission of off-gas does not take place at a constant rate, the sharply fluctuating emission rates can be attributed to the circumstance that, during some sampling, off-gas was being

given off, but in others, it was not.

Table 7 shows the mean percentage, determined from several separate measurements (usually 9), of the CO_2-bound C-14 in the effluent gas from the various pressurized-water reactors.

Table 7. Mean percentage of CO_2-bound C-14 in the total C-14 emission from pressurized-water reactors

Nuclear power plant	Period	Mean percentage of CO_2-bound C-14 in the total C-14 activity in the effluent gas	Mean value from
Obrigheim	1977	13%	9 random samples
	1978	57%	9 random samples
	1978	55%	4 weekly samples
Stade	1978	30%	2 monthly samples
Biblis A	1977	9%	8 random samples
	1978	30%	9 random samples
Biblis B	1977	3%	9 random samples
	1978	9%	9 random samples
Neckarwestheim	1977	3%	9 random samples
	1978	5%	9 random samples

4.3.2. Measurements of the C-14 concentration in the effluent gas

(i) The nuclear power plant, Obrigheim

At the nuclear power plant, Obrigheim, samples of the effluent gas were taken at about monthly intervals throughout 1977 and 1978 and the proportions of CO_2-bound, CO-bound and hydrocarbon-bound C-14 were determined separately. The total C-14 emission in the stack effluent gas lay in the range 2.10^{-10} to $1,4.10^{-8}$ Ci/m^3. The mean value for 1977 amounted to $2,9.10^{-9}$ Ci/m^3, and for 1978 to $8,1.10^{-10}$, whence, using the air

throughput of 120 000 m^3/h, an annual emission of 3,0 Ci in 1977 and 0,9 Ci in 1978 is found.

The different percentages of CO_2-bound C-14 for the 2 years are striking. In 1977 the mean percentage of CO_2-bound C-14 was 13%, but in 1978 57%. The emission of CO_2-bound C-14 is about the same for each year: 0,4 Ci/a in 1977 and 0,5 Ci/a in 1978.

Since June 1978 there has also been continuous separation of all the C-14 compounds. The results of these measurements agree well with those of the random samples made in parallel (mean C-14 concentration $9{,}0.10^{-10}$ Ci/m^3, mean percentage of CO_2-bound C-14 61%). From the annual emission the normalized annual emission relative to the reactor capacity is calculated to be 8,7 Ci/GWa for 1977 and 2,5 Ci/GWa for 1978. Relative to the actual power generated, these values are rather higher: 11,6 Ci/GWa in 1977 and 3,0 Ci/GWa in 1978. The individual results are collected together in Tables A11, A12, A13 and Figs. A7, A8 in the Appendix.

(ii) The nuclear power plant, Stade

Since April 1977 continuous separation of the CO_2 component in the stack gas at the nuclear power plant Stade has been carried out (monthly total samples).

Since September 1978 continuous, complete separations of the C-14 have been carried out. The 2 monthly-total samples so far available show quite different CO_2 percentages. In the sample taken in September 1978 the CO_2 fraction was 53%; in the October sample 7% was measured.

In these two years the concentration of CO_2-bound C-14 lay in the range 3.10^{-10} to $10.^{-8}$ Ci/m^3. The highest concentrations (May 1977 with $1{,}4.10^{-8}$ Ci/m^3 and April 1978 $3{,}3.10^{-9}$ Ci/m^3) occured due to the periods for overhaul and change of fuel elements.

With an air throughput of 150 000 m^3/h, the emission per month calculated from the concentration values is 0,03 to 1,3 Ci. For 1977 an emission of CO_2-bound C-14 of 3 Ci was calculated, and for 1978 an emission of 1,5 Ci. It is to be noted here that the highest value measured, viz., $1,4.10^{-9}$ in May 1977, contributed essentially to the difference between the two years. Since only 2 monthly samples with separation of the C-14 compounds are so far available for the Stade nuclear power plant, we do not attempt to estimate the total emission of C-14. Relative to the reactor capacity, the normalized annual emission of CO_2-bound C-14 came to 4,5 Ci/GWa in 1977 and 2,3 Ci/GWa in 1978. Relative to the actual power generated, the mean annual emission for CO_2-bound C-14 amounted to 4,8 Ci/GWa in 1977 and 3 Ci/GWa in 1978. The individual results are collected together in Tables A14, A15, A16 and Figs. A9 and A10 in the Appendix.

(iii) The nuclear power plant Biblis Block A

The separate measurements carried out at intervals of about a month at the nuclear power plant Biblis Block A gave C-14 concentrations in the effluent gas between $3,4.10^{-10}$ and $9,4.10^{-9}$ Ci/m^3. The mean total C-14 concentration amounted to $3,4.10^{-9}$ Ci/m^3 in 1977, and $1,5.10^{-9}$ Ci/m^3 in 1978, from which, using the air throughput of 160 000 m^3/h, the annual emission came to 4,8 Ci in 1977 and 2,1 Ci in 1978. The mean CO_2-bound fraction of the total emission was 9% in 1977 and 32% in 1978. These give annual emissions of CO_2-bound C-14 of 0,4 Ci in 1977 and 0,6 Ci in 1978. Here too the CO_2-bound percentages in the individual measurements are very variable; values between 3 and 85% of CO_2-bound C-14 were determined. The normalized annual emission relative to the reactor capacity was calculated to be 4,0 Ci/GWa for 1977 and 1,7 Ci/GWa in 1978; relative to the actual current generated, these values are rather higher, namely, 6,4 Ci/GWa in 1977 and 2,0 Ci/GWa in 1978. The individual results are collected in Tables A17, A18, and Figs. A11 and A12 in the Appendix.

(iv) The nuclear power plant Biblis Block B

In the effluent gas from the nuclear power plant Biblis Block B both random sample measurements (at intervals of about a month) and continuous separation of the CO_2-bound C-14 were carried out. For the evaluations the CO_2 data obtained from the continuous separation and the monthly determinations were used, and the total emission was calculated from the proportions of the individual forms of compounds determined in the monthly random samples. In 1977 the concentration of CO_2-bound C-14 lay in the range from 5.10^{-12} to $2,1.10^{-10}$ Ci/m^3, with a mean value at $8,8.10^{-10}$ Ci/m^3. The proportion of CO_2-bound C-14 lay in the range from 0,1 to 10%, with a mean value at 2,5%. Taking into account the air throughput, the mean values give an annual emission of 4,9 Ci, of which 0,12 comes from the CO_2-bound C-14. In 1978 with $3,4.10^{-9}$ Ci/m^3, approximately the same mean C-14 concentration as in 1977 was determined. This corresponds to an annual emission of 4,8 Ci. The proportion of CO_2-bound C-14 was rather larger at 9% than in 1977; this corresponds to an annual emission of CO_2-bound C-14 of 0,42 Ci. From the annual emissions, the normalized annual emissions relative to the actual power generated are calculated to be 5 Ci/GWa for 1977 and 7,2 Ci/GWa for 1978.

However, the 1978 calculations based solely on the measurements of the random samples give values lower by about 30%, namely, a mean C-14 concentration of $2,4.10^{-9}$ Ci/m^3, an annual emission of 3,4 Ci for all the C-14, and an annual emission of 0,29 Ci of C-14 in the CO_2-bound form. The individual results are collected together in Tables A19, A20, A21 and Figs. A13, A14, and A15 in the Appendix.

(v) The nuclear power plant, Neckarwestheim

At the nuclear power plant at Neckarwestheim, starting from April 1977, samples were taken at about monthly intervals, and the concentrations of the CO_2-bound, CO-bound, and hydrocarbon-bound fractions of the C-14 were determined separately. From August 1977, additionally, continuous separation of CO_2 in the stack gas has been carried out.

In 1977 the period over which the continuous separation was carried out was too short to obtain the annual emission from these results. But from the random sample measurements the C-14 concentration lay in the range $1{,}4.10^{-10}$ to $5{,}0.10^{-9}$ Ci/m^3. With a mean value of $2{,}3.10^{-9}$ Ci/m^3; from these values and the air throughput of 200 000 m^3/h an annual emission of 4 Ci is obtained for 1977, and with a mean CO_2-bound fraction of 2,7%, an annual emission of 0,11 Ci from the CO_2-bound C-14. In 1978 the random sample measurements gave a mean C-14 concentration of $1{,}8.10^{-9}$ Ci/m^3 and a proportion of 5% of CO_2-bound C-14. These values give an annual emission of 3,2 Ci for 1978, of which 0,15 Ci comes from the CO_2-bound C-14.

Somewhat higher values are obtained from the measured results of the continuous CO_2 separation and the CO_2-bound percentage for 1978. From these measurements a mean C-14 concentration of $2{,}2.10^{-9}$ Ci/m^3 and an annual emission of 3,9 Ci were determined for 1978; of the latter 0,16 Ci was due to the CO_2-bound C-14. Since the latter data are statistically more reliable, these values were put into the summary table. Relative to the actual power generated, we obtain from these values annual emissions of 6,6 Ci/GWa for 1977 and 6,3 Ci/GWa for 1978. The individual results are collected together in Tables A22, A23, A24 and Figs. A16, A17, A18 and A19 in the Appendix.

4.3.3. Summary of the measured results for pressurized-water reactors

The annual emissions of C-14 determined in the effluent gas from pressurized-water reactors and the annual emissions per GWa of current generated are shown together in Table 8. It is to be noted, in general, that for pressurized-water reactors the annual emissions should be determined for accuracy by a multiplicity of measurements over a period of about 2 years. Also, the marked fluctuations observed in the individual measurements, particularly of the hydrocarbon-bound fraction of C-14, are smoothed out by averaging over a sufficiently large number of individual values and should give representative

values. It is noteworthy that even the mean value of the separate C-14 compounds can vary considerably from one plant to another, and also at the same plant from one year to another. From the results to date it seems that a higher proportion of CO_2-bound C-14 is present in the effluent from older installations than for newer installations. In the calculation of the annual emission relative to the power generated in 1978, it is to be noted that only the power generated for the first half-year was known, and so the value for the whole year was calculated on the assumption that the circumstances would be the same in the second half-year.

Table 8. Synopsis of the emission of C-14 in the effluent gas from pressurized-water reactors

Nuclear power plant	Year	Annual emission	Annual emission relative to the power generated
		Ci	Ci/GWa
Obrigheim	1977	3,0	11,6
	1978	0,9	3,0
Stade [1]	1977	3,0	4,8
	1978	1,5	3,0
Biblis A	1977	4,8	6,4
	1978	2,1	2,0
Biblis B	1977	4,9	5,0
	1978	4,8	7,2
Neckarwestheim	1977	4,0	6,6
	1978	3,9	6,3

1) Only the CO_2-bound C-14 measured.

Averaging the annual emission per GWa generated for the several installations gives an annual emission of

6 Ci/GWa

for pressurized-water reactors. This corresponds to a mean annual emission of 5 Ci for a pressurized-water reactor of 1000 MW installed electrical capacity.

4.4. C-14 measurements in extraction ducts of a boiling-water reactor and of a pressurized-water reactor

In order to be able to draw conclusions as to the contributions of the various areas in a nuclear power plant to the total amount of C-14 emitted in the stack effluent, measurements, over several days in each case were made in the separate branch lines of the air extract system of a boiling-water reactor (the nuclear plant at Brunsbüttel) and of a pressurized-water reactor (the Neckarwestheim plant). It was established for both types of reactors that the main part of the C-14 emission takes place via the off-gas from the off-gas section. The other C-14 emissions come from all parts of the equipment in the primary circuit or from sub-sections of the auxiliary equipment building in which the primary cooling medium is purified.

(i) The nuclear power plant at Brunsbüttel

From June 14th to 16th,1978 the C-14 activity concentrations were measured in the separate branch lines and in the total stack effluent at the Brunsbüttel nuclear plant, and from the amounts of exhaust air in these branch lines the rates of discharge from the various areas were determined. The main contribution, 42%, to the total emission during the period was from the off-gas system, 29% from the turbine hall, 11% from the reactor building and auxiliary building; 18% of the total emission (100% in the stack) was not recorded. No measurements were carried out in the exhaust air from the interspace or the pump seal extracts, or the control area of the operations building. Because of the special operating conditions at the Brunsbüttel nuclear power station, however, before and during the C-14 sampling, these results should not be generalized. The individual results are collected together in Tables A25 and A26 in the Appendix.

(ii) The nuclear power plant at Neckarwestheim

A similar series of measurements was carried out at the Neckar-

westheim nuclear power plant from August 14th to 16th, 1978 the C-14 was separately determined in the CO_2, CO, and hydrocarbon forms. In this period about 84% of the total emission occurred via the reactor building (8% from the ring rooms, 76% from the operational rooms), and about 16% from the auxiliary equipment plant. The highest C-14 concentrations have been measured in off-gas but none was released during the period examined here. By comparing the emission rate determined in the stack effluent during the sampling when no off-gas was given off, which corresponds to an annual emission of 1,1 Ci, with the annual emission as determined by the continuous measurements, which came to 3,9 Ci for 1978, it was estimated that about 60 to 70% of the C-14 discharged is emitted via the off-gas.

In the individual branches very different proportions of the individual C-14 compounds were measured, lying in the range from 0,7% of CO_2-bound C-14 in the off-gas up to 54% in the operation rooms. The individual results are collected together in Tables A27, A28, and A29 in the Appendix.

4.5. Measurement of C-14 compounds in the effluent gas from the reprocessing plant at Karlsruhe (WAK)

The C-14 emission from WAK has already been determined in previous investigations by the Nuclear Research Centre, Karlsruhe (SCH 77/1, SCH 77/2, SCH 77/3). These measurements were restricted to the emission of CO_2-bound C-14. In discussions on the problem of retaining the C-14 at large reprocessing plants, the question arose whether other forms of compound appear as well as the CO_2-bound C-14, and if so, in what proportions. To clarify this question we carried out measurements of the C-14 in the effluent gas from WAK during a dissolution cycle; the C-14 was measured separately in the form of CO_2, CO, and hydrocarbon compounds.

The sampling took place after the aerosol filter in a branch line from the stack effluent. With a throughput rate of 0,6 to

0,7 litre per minute the effluent was passed through a bubbler containing 200 ml of Naoh, so that the CO_2 compounds were completely separated.

After passing through the bubbler, the effluent was fed into a test gas holder, since separation of CO-bound and hydrocarbon-bound C-14 in situ would have led to too much cost.

The sampling took place continuously over 19 hours, in stages of 1 hour or 2 hours starting at the hour. The first sample was taken about 24 hours after the start of the current dissolution of the fuel material. The mechanical chopping of the fuel began at 5.45 p.m. on April 19th, 1978; the chemical dissolution of the fuel began at 7.45 p.m.

The measurements showed that the proportions of compounds such as CO or hydrocarbons is negligibly small. The total proportion of such compounds amounted to 0,12% of the total emission, i.e. 99,88% of the C-14 is emitted as CO_2. Slight quantities of CO and hydrocarbons could be detected starting from the mechanical chopping of the fuel elements up to 8 hours after this process. The highest proportion of these types of compound amounted to 0,28% in the first hour after the chemical dissolution of the fuel.

The individual results are collect ed together in Tables A30, A31, A32 and Figs. A20 and A21 in the Appendix.

These measurements confirm that, as stated in section 2.5, measurements of the CO_2-bound C-14 include, within the limits of measurement error, the total emission of C-14.

5. Radiation exposure

5.1. Models for calculation of the dose

The radiation exposure by C-14 intake may be calculated using a specific activity model. Two methods are distinguished in the literature:

1) the specific activity of the carbon in the human body, in tissue, or in an organ is equated to the specific activity of the carbon in the atmosphere;
2) the specific activity of the carbon in vegetation is equated to the specific activity of carbon in the atmosphere. The dose is then calculated from the intake of C-14 by inhalation of contaminated air and by ingestion of contaminated foodstuffs.

In the first case, in order to calculate the dose, one needs the effective energy of the C-14 for the specific organ, the mass of the organ, and the mass of the carbon in the organ. If the second method is used, then, in addition to the physiological parameters which are included in the dose factor, certain other parameters enter into the calculation of the radiation exposure. These include, for example, the habitual consumption and the transfer factors which describe the transfer of carbon-14 from fodder into animal products.

When values of the radiation exposure for the whole body are given in this text, they are always to be understood in the sense of the total-body dose as defined in ICRP-2. For C-14 these total-body doses are equal in value to the effective dose in the sense of ICRP-26, as can easily be shown using the dose factors from Table 9 and the weight factors in ICRP-26 for determining the effective dose. For this reason the total-body doses given in this report can also be interpreted as effective doses.

5.1.1. Model 1

The dose rate D in rem/a is given by the relation

$$D = 1{,}87.10^{10}.A_s.m.E_{eff}/m_o,$$

where

A_s = specific activity of the carbon in the organ, in Ci/g

m = mass of carbon in the organ, in g,

E_{eff} = effective energy in the organ, in MeV,

m_o = mass of the organ, in g.

The dose factor, i.e., the quotient D/A_s, is found from the above equation, in the units rem/a per Ci/g. These factors were calculated by Killough and Rohwer and are given in Table 9. The dose factors in Table 9 were calculated using the physiological data (according to ICRP-23) for a male adult. It is assumed here that these values do not depend significantly on a person's age.

Table 9. Dose factors for C-14 according to Killough and Rohwer (KIL 78/1)

Organ	Dose factor ((rem/a)/(Ci/g))
Whole body	$2{,}1.10^8$
Skeleton:	
Endosteal cells	$3{,}3.10^8$
Red bone marrow	$3{,}7.10^8$
Yellow bone marrow	$5{,}8.10^8$
Bones	$1{,}2.10^8$
Lungs	$9{,}1.10^7$
Liver	$1{,}3.10^8$
Kidneys	$1{,}2.10^8$
Spleen	$1{,}0.10^8$
Thyroid gland	$9{,}5.10^7$
Testicles	$8{,}0.10^7$
Intestinal tract	$1{,}6.10^8$
Adipose tissue	$6{,}2.10^8$

5.1.2. Model 2

The contamination of plants by C-14 is given by

$$C_{pf} = C_L . f_c / m_c \ ,$$

where C_{pf} = concentration of C-14 in the plants, in Ci/kg
C_L = concentration of C-14 in the air, in Ci/m^3
f_c = fraction of natural carbon in total mass of plant
m_c = mass of carbon in kg per m^3 of air.

The carbon-14 content of animal foodstuffs can be determined from C_{pf} by using the relation

$$C_j = C_{pf} . I . T_{pf,j} \ ,$$

where

C_j = concentration of C-14 in the animal product j, in Ci/kg,
C_{pf} = concentration of C-14 in the plants, in Ci/kg,
I = animal's fodder intake, in kg wet weight/d,
$T_{pf,j}$ = transfer factor from animal fodder to the animal product j, in d/kg or d/l.

The dose rate in an organ is then given by

$$D_{i,a} = g_{i,a} \sum_j C_j . U_{j,a} \ ,$$

where

$D_{i,a}$ = dose rate in organ i for the age-group a, in rem/a
$g_{i,a}$ = dose factor for organ i and age-group a, in rem/Ci
$U_{j,a}$ = amount of food eaten per year of vegetable or animal products of category j by a member of the age-group a, in kg/a,
C_j = C-14 concentration in food of catgeory J, in Ci/kg.

The method just described of calculating the radiation exposure due to C-14 is used in the USRNC Guide 1.109 (Rev. 1) (NRC 77) and in the "General bases of calculations for determining the radiation exposure due to the emission of radioactive materials in effluent gas" (BMI 77). The data used in these papers for calculating the radiation exposure in accordance with the above formulae are compared in Table 10. The symbols used in the first column have the meanings already defined.

Table 10. Data for calculating the radiation exposure, as given in (NRC 77) and (BMI 77)

Symbol	Unit	Meaning		Values given in (NRC 77)	(BMI 77)
f_c	-	fraction of carbon in total plant mass		0,11	0,11
m_c	kg/m^3	mass of carbon in the atmosphere		$1,6.10^{-4}$	$1,8.10^{-4}$
I	kg/d	moist weight of fodder intake:	cattle	50	55
			goats	6	-
$T_{pf,j}$	d/kg	fodder to meat transfer factor		0,031	0,031
		fodder to milk transfer factor:	cattle	0,012	0,012
			goats	0,10	-
$U_{j,a}$	kg/a	maximum food intake:			
		vegetables products (greens):	adult	584	462
			adolescent	672	-
			child	546	-
			baby	-	-
		milk:	adult	310	330
			adolescent	400	-
			child	330	-
			baby	330	300
		meat:	adult	110	150
			adolescent	65	-
			child	41	-
			baby	-	-
$g_{i,a}$	rem/Ci	ingestion dose factor:			
		whole body:	adult	568	630
			adolescent	812	-
			child	2420	-
			baby	5060	4810
		liver, thyroid, spleen, lungs, intestinal tract:	adult	568	630
			adolescent	812	-
			child	2420	-
			baby	5060	4810
		bones:	adult	2840	3800
			adolescent	4060	-
			child	12100	-
			baby	23700	4810
		kidneys:	adult	568	630
			adolescent	812	-
			child	2420	-
			baby	5060	630
		adipose tissue	adult	-	750
			adolescent	-	-
			child	-	-
			baby	-	-

5.1.3. Comparison of Models 1 and 2

From the data given in Table 10 dose factors which give the annual dose per unit specific activity of carbon in the atmosphere can also be calculated. The results of such a calculation are shown in Table 11. For comparison the dose factors given by Killough and Rohwer (KIL 78/1), which are obtained by using the first method, are also shown in Table 11. The values from the 1977 UNSCEAR report (UNSCEAR 77) are also quoted.

Table 11: Dose factors in mrem/a per (pCi (C-14) / (g (C-12))

D A T A	S O U R C E S			
	(BMI 77)		(KIL 78/1)	(UNSCEAR 77)
Organ	adults	children	adults	adults
whole body	0,065	0,11	0,21	0,21
bones	0,40	0,11	0,12	-
periosteum	-	-	0,33 [1)]	0,33
red bone marrow	-	-	0,37	0,36
gonads	-	-	0,080 [2)]	0,082
liver	0,065	0,11	0,13	-
lungs	0,065	0,11	0,091	-
kidneys	0,065	0,012	0,12	-
adipose tissue	0,078	-	0,62	-

1) endostial cells. 2) testicles.

Comparison of the dose factors in Table 11 from (KIL 78/1) with those from (BMI 77) shows that the first method (Model 1) gives higher values in general than the second method (Model 2).

In Model 1 it is assumed that the specific activity of the carbon in an organ is equal to the specific activity of carbon in the atmosphere. Consequently this method gives the highest conceivable concentration of C-14 in the organs, and hence leads to the highest conceivable radiation exposure. Measurements e.g. (KRU 70), (OSM 75), (YEM 69) show that the specific activity can be changed by 5% due to the isotope effect in the general exchange of carbon.

In the second method it is merely assumed that the specific activity of the carbon in the atmosphere is in equilibrium with the carbon in plants.

5.2. Local radiation exposure

The place-dependent specific activity of the carbon in the atmosphere near the ground in the vicinity of a nuclear power plant is given by the formula

$$A(x) = Q.\chi(x)/m_c ,$$

where

A(c) = specific activity at distance x from the stack of the nuclear power plant in (Ci(C-14))/(g(C-12))

Q = strength of the emission source in Ci/S,

$\chi(x)$ = meteorological dispersion factor, in s/m^3,

and m_c = carbon content in the air, in g/m^3.

If we make the conservative assumption (as, for example, in (BMI 77)) that a man is situated at a point of maximum C-14 concentration in the atmosphere, and consumes only foodstuffs produced at this point, then the dose is proportional to the corresponding meteorological dispersion factor. If we choose a dispersion factor of 3.10^{-7} s/m^3, which can be regarded as representative for the most unfavourable place in the vicinity of a nuclear power plant with a 100 m high stack, then the maximum values of the radiation exposure due to C-14 are as shown in Table 12. In the calculation the dose factors given in the first two columns of Table 11 are used.

Table 12. Radiation exposure due to C-14 at the place of maximum effect in the vicinity of a nuclear power plant under standard dispersion conditions (see text)

Type of reactor	Rate of emission 1)	Proportion of CO_2 1)	Maximum radiation exposure in mrem/a for		
			bones adult	whole body adult	whole body baby
BWR	10 Ci/a	100%	0,2	0,03	0,06
PWR	5 Ci/a	10%	0,01	0,002	0,003

1) These rates of emission and proportions of CO_2 are valid for a nuclear power plant with nominal capacity 1000 MW.

A more realistic estimate of the radiation exposure by ingestion is obtained if the catchment area from which the food intended for consumption comes is taken into account. Kelly et al. (KEL 75) assume, for example, that 50% of the food is produced within a circle of radius 50 km, while the remaining food is essentially uncontaminated. Under this assumption, according to (KEL 75), the values of the radiation exposure at the position with maximum airborne concentrations in the vicinity of the site are reduced by a factor of 20.

For pressurized-water reactors only the $^{14}CO_2$ contribution is considered for the exposure by ingestion, because only this compound is taken up by the assimilation process of plants and therefore enters into the food chain. This method of approach is justified in the close vicinity of a nuclear power plant because the time of transportation of the hydrocarbon forms of C-14 to the observation point is small compared with the time needed for the oxidation of hydrocarbon-bound C-14 to $^{14}CO_2$ in the atmosphere. The time of transportation of the emitted C-14 from the source to the point of maximum airborne concentration is of the order of magnitude of about 10 minutes; but, according to (LEV 78), methane persists in the atmosphere for 6 to 7 years on average.

For dose by inhalation, on the other hand, the total discharge of C-14 from pressurized-water reactors is taken into account.

To the radiation exposure values shown in Table 12 for the infant whole body, inhalation of contaminated air and ingestion of contaminated milk contribute respectively 1% and 99% in the case of boiling-water reactors, and 10% and 90% in the case of pressurized-water reactors. Table 13 shows the contributions, via the exposure pathways considered, to the total radiation exposure of an adult.

In the calculation of the local radiation exposure the discharge data and meteorological data which correspond to the growing season should be used in the calculation of the dispersion. Killough and Rohwer have shown by an example that even for a uniform discharge rate the dose could be under-estimated

Table 13: Contributions by various exposure pathways to the radiation exposure of an adult

Exposure pathway	contribution to dose	
	BWRs	PWRs
Inhalation	1 %	6 %
Ingestion of:		
vegetable products	45 %	42 %
green vegetables	4 %	4 %
milk	23 %	22 %
meat	27 %	26 %

by a factor of 3 if this correction of the meteorological data is not made.

The values of the radiation exposure given in Table 12 correspond to the increase in the specific activity of carbon in the atmosphere and vegetation at the place most adversely affected in the vicinity of a boiling-water reactor of 0,5 pCi/g, or, in the case of a pressurized-water reactor, an increase of 0,3 pCi/g, taking into account the proportion of C-14 which is present in the form of hydrocarbons. 100 years ago the specific activity of carbon amounted to some 6 pCi/g. Due to the testing of nuclear weapons up to 1963 the specific activity was about doubled in the troposphere, and it amounts at present to about 8 pCi/g. In view of the accuracy of C-14 measurements it can be expected that the local increase of the specific activity can be detected by the measuring techniques. Investigations to do this have already been carried out in various places. Münnich has shown that local depressions due to the combustion of fossilized carbon materials must be taken into account in the measurement of C-14 in the vicinity of a nuclear power plant (MÜN 78).

5.3. Estimation of the regional exposure

Estimation of the regional exposure due to the emission of carbon-14 in the effluent gas from nuclear power plants presents difficulties. This circumstance is due principally to

the fact that the generally available dispersion models which describe the atmospheric distribution of the emitted carbon provide reliable data only up to distances of 10 km to 20 km from the place of emission. For greater distances the difficulty consists in bringing the changing meteorological conditions into the calculation. For this reason we give here only the orders of magnitude within which the dose lies at greater distances from the point of emission. Kelly et al. (KEL 75) give the dose for a range of 1 km to 1000 km from the plant in question. For a distance of 1 km one has to reckon on having the maximum radiation exposure, as calculated in section 5.2. According to Kelly, this value falls by about 3 orders of magnitude up to a distance of 1000 km. Moreover, for distances up to 1000 km the hydrocarbon-bound C-14 should still not make any essential contribution to the radiation exposure, because the time of transportation in the atmosphere, about 10 days, is slight compared with the average persistence time of hydrocarbons, which is in the region of several years.

5.4. The global radiation exposure

The carbon-14 emitted from nuclear engineering installations is distributed worldwide in the atmosphere and biosphere until, in the course of centuries, it will be deposited in the depths of the sea. To estimate the global radiation exposure one generally calculates the collective dose commitment in man-rem relative to the total world population. Because of the long half-life of C-14 and its behaviour in the environment, the emission of radioactive carbon leads to a long period of contamination of the environment. This time period must be taken into account in determining the effects on people of emissions of carbon-14, i.e., in calculating the collective dose commitment. There are already several publications in which the biosphere is described by multi-compartmental models. Generally the stratosphere, the troposphere, the terrestrial biosphere, the upperlayers of the oceans and the sea deeps are regarded as the compartments, but in some models a more detailed sub-division into compartments is adopted.

The collective dose commitment is proportional to the time-integral of the product of the world population and the specific activity of the carbon in the atmosphere. In the older literature the question is left open whether the time interval for calculating the collective dose commitment should be from a few decades or a century, or whether the integration should be carried out for $t \to \infty$.

The time dependence of the mean dose commitment due to the emission of 1 Ci of C-14 is shown in Fig. 6.

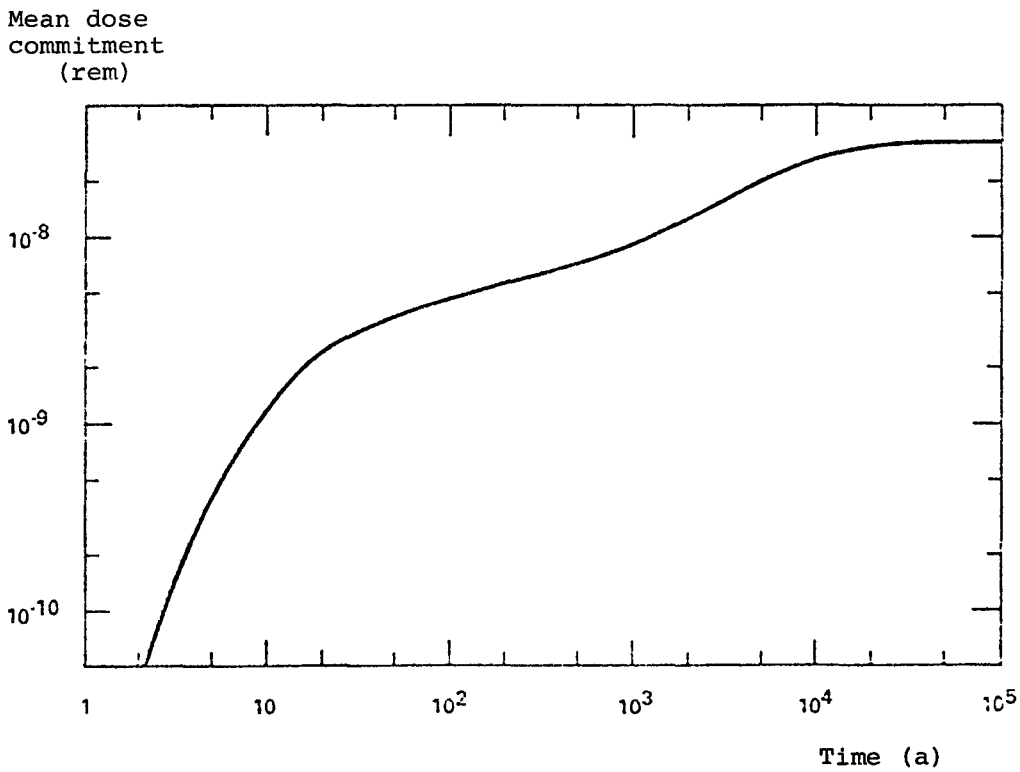

Fig. 6. Time dependence of the mean dose commitment in the latitude range between 30° and 90° due to the emission of 1 Ci of C-14 during a year (according to (BER 77/1))

It can be seen from Fig. 6 that, assuming a constant world population, the collective dose commitment reaches its asymptotic, limiting value after about 20,000 years.

In the more recent literature (e.g., (KIL 78/2)) it becomes increasingly apparent that no arbitrary time limit can be set

in estimating the collective dose commitment, i.e., the integration is to be carried out for $t \longrightarrow \infty$. The effect of the combustion of fossilized carbon on the specific activity has been studied by several authors. Despite the fact that forecasts of the energy needs within the time interval to be considered are very uncertain it appears that the specific activity will not be reduced significantly by the combustion of fossilized carbon. So this effect leads to no essential reduction in the radiation exposure due to C-14, and it can therefore be neglected in estimating the collective dose commitment.

It has already been pointed out in Section 5.2 that the mean period of persistence of hydrocarbon-bound C-14 in the atmosphere before oxidation to CO_2 occurs is of the order of several years. Moreover, Fig. 6 shows that the carbon-14 present in the atmosphere makes only a slight contribution during the first few years to the collective dose commitment. Therefore in calculating the collective dose commitment the hydrocarbon-bound C-14 can be treated as if it were CO_2.

Killough and Till bring into their calculations of the collective dose commitment the future expansion of the world population as estimated by the United Nations Organization in 1974 (UN 74). According to this forecast an increase of the world population up to $12,21.10^9$ persons in 2075 is calculated. After 2075 the world population is assumed to be constant. Bergman and McEwan assume in their calculation that the world population will not exceed 10^{10} persons (BER 77).

Killough and Rohwer have standardized some of the values of the collective dose commitment given in the literature, using the (UN 74) forecast of the expansion of the world population. These values, with 2 new additional ones, can be found in Table 14.

It follows from the values in Table 14 that due to the emission of 1 Ci of carbon-14 into the atmosphere a collective dose commitment of about 500 man-rem is to be reckoned on. During the first 100 years a collective dose commitment of about

Table 14. Collective dose commitment due to the emission of 1 Ci of C-14 into the atmosphere (range of integration extending to t $\longrightarrow \infty$)

Source	Collective dose commitment (man-rem)
(UNSCEAR 1972)	520
(USERDA 1975)	530
(MAR 74/1)	480
(KEL 75)	400
(BRO 74)	590
(KIL 78/1)	540
(BON 77)	490
(BER 77)	320

50 man-rem is to be reckoned on (see Fig. 6).

5.5. Evaluation

A comparative evaluation of the radiation exposure of the population of the European Community due to the emission of krypton-85, tritium, carbon-14, and iodine-129 by the nuclear industry up to the year 2000 was made in the report (KEL 75). In (WHO 78) the collective dose commitment of the population per MWa of electrical energy generated by nuclear power plants is given. Here too it appears that the contribution to the radiological exposure of the population due to C-14 is substantially important even if a 90% retention at the discharge point of the C-14 is assumed. In (SCH 76) the radiation exposure due to C-14 from nuclear installations is compared with the radiation exposure due to the C-14 occurring in nature. In the report (USEPA 76) the effects due to the emission of C-14 from nuclear establishments are evaluated as regards possible health hazards to the population.

In the previous sections methods of calculation for the determination of the local, regional, and global radiation exposure due to C-14, and their results have been given. The questions now arise as to the relative significance of the local and global

radiation exposure due to C-14, and the evaluation of the radiation exposure due to nuclear plants in comparison with the natural radiation exposure due to C-14. As a basis for the evaluation, the risk of somatic damage to an individual may be used, for example; these values have been assembled in Table 15. The calculations were carried out using the risk factors for C-14 given in (KIL 78/2), which in turn are based on the values in (ICRP 26).

Table 15. Comparison of the risk of a man's developing cancer as a result of radiation exposure due to C-14

	Risk
Person living 70 years at the point of maximum effect in the vicinity of a boiling-water reactor (data as in Table 12)	2.10^{-7} 1)
Person living 70 years at the point of maximum effect in the vicinity of a pressurized-water reactor (data as in Table 12)	1.10^{-8} 1)
Person who will be exposed from the year 2005 to 2075 to the global radiation exposure due to:	
a) C-14 discharged into the atmosphere as a result of the use of atomic energy for generating electrical power	1.10^{-7} 2)
b) the nuclear weapons tests from 1945 to 1974	1.10^{-6}
c) the C-14 naturally present	8.10^{-6}

1) Here only the radiation exposure local to the reactor is taken into account

2) Development of energy consumption and of retention techniques according to (KIL 78/2), from which the "intermediate scenario" has been taken

A comparison of the numerical values in Table 15 shows that an individual's cancer risk at the point of maximum effect in the vicinity of a reactor corresponds to that which can be deduced from the global distribution of C-14 in the atmosphere in the next 100 years due to the peaceful use of nuclear energy. In this connection it is to be noted that the local radiation exposure exists only while the nuclear plant is operating, while the global radiation exposure after contamination of the

biosphere has taken place extends over a period of several thousand years.

Calculations of the radiation exposure due to C-14 are based on the specific activity of the carbon in the atmosphere near the ground. This specific activity approach leads to realistic results in the calculation of the global radiation exposure, since in this calculation a largely homogeneous distribution of C-14 can be assumed. In calculating the local radiation exposure, however, the specific activity at the point of maximum effect in the vicinity of the nuclear power plant was introduced. This approach leads to an over-estimate of an individual's dose, since it cannot generally be assumed that the whole of a person's food will come from the point of maximum effect.

6. Concluding remarks

The results referred to in this report of investigations and studies into the emission of carbon-14 from nuclear engineering establishments, its measurement, and distribution in the environment, and the resulting radiation exposure are based on the evaluation of more than 60 scientific publications and more than 700 of our own measurements. In general a satisfactory agreement is found between the theoretical calculations of the rates of formation and liberation and the measurements of emissions at the nuclear power plants. Also there is reasonably agreement between the various published calculations of the local radiation exposure and the long term global collective dose commitment to be expected.

Calculations and measurements show that, radiologically speaking, carbon-14 is one of the most important nuclides in nuclear engineering. Its relative significance has increased, since the emission of other long-life radioactive nuclides has been reduced by improved retention techniques.

The contribution of carbon-14 from a large light-water reactor of modern design to the local radiation exposure lies in the range 0,01 to $\lesssim$ 1 millirem per annum. A slight increase of the C-14 level in the vicinity of a nuclear engineering plant can, in principle, be detected by sensitive measuring techniques. This local increase is, however, smaller than the range of local fluctuations of the specific activity of carbon compounds produced by the natural formation of carbon-14 in the atmosphere.

Because of the long half-life and the worldwide distribution of carbon-14, the long-term global effects have also to be considered; a measure of these effects is the long-term collective dose commitment occurring globally. However, to reduce these contributions, carbon retention techniques can be put into operation, and these are already envisaged for future reprocessing plants.

Through the investigations made it has been possible to

complete essentially the data on the emission of C-14 from light-water reactors, so that for these reactors adequately confirmed data are now available for reference.

In contrast, there are fewer experimentally confirmed data for other types of reactor. Investigations with gas-cooled reactors and reactors moderated by heavy water would therefore be desirable.

For such investigations it would be possible essentially to fall back on known methods of sampling and measurement. The interest lies in putting into operation simple methods for routine use. This has largely been solved by the adoption of continuous sampling methods for CO_2-bound C-14 and measurement in liquid scintillation counters. The development and adoption of solid adsorbent materials would be desirable to simplify the sampling. In the German Federal Republic it is envisaged that, in the surveillance of light-water reactors, continuous separation of the CO_2 in the effluent gas will be carried out, together with a C-14 balance in a mixed sample every quarter year.

For a better understanding of the liberation pathways within a nuclear plant and for estimating the separate contributions to the total emission at nuclear power plants it is of concern to know in which processes of the operation and via which parts of the plant the emission of C-14 takes place. The initial basic data in such a survey have been obtained by measurements at two plants, but further investigations are necessary for this purpose.

Measurement of the C-14 content of environmental samples in the environment close to nuclear power installations would also be of interest, in order to confirm the calculations and to check the dispersion calculations if possible.

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APPENDIX

Compilation of individual results

In cases where C-14 concentrations are calculated using discontinuous samples, in the following tables, minor deviations may occur between monthly and annual mean values. These are caused by several C-14 measurements performed during one month, being given equal weighting in the annual mean value.

Table A1.: Concentration of CO_2-bound C-14 in the effluent gas from the Gundremmingen nuclear power plant during normal reactor operation

Year	Sample taken	C-14 concentration (Ci/m^3)
1976	29.10.	$1,2 \cdot 10^{-9}$
	12.11.	$2,8 \cdot 10^{-9}$
	19.11.	$1,0 \cdot 10^{-9}$
	16.12.	$2,3 \cdot 10^{-9}$
	21.12.	$1,1 \cdot 10^{-9}$
	29.12.	$1,0 \cdot 10^{-9}$
1977	05.01.	$1,5 \cdot 10^{-9}$
	12.01.	$4,0 \cdot 10^{-9}$

Table A2: Concentration of CO_2-bound C-14 in the effluent gas from the Gundremmingen nuclear power plant after shut-down (monthly mean values)

Year	Sample taken	C-14 concentration (Ci/m^3)
1977	January	$2,3 \cdot 10^{-9}$
	February	$5,4 \cdot 10^{-10}$
	March	no sample taken
	April	no sample taken
	May	$4,5 \cdot 10^{-10}$
	June	$3,0 \cdot 10^{-10}$
	July	$1,3 \cdot 10^{-9}$
	August	$3,1 \cdot 10^{-10}$
	September	$9,7 \cdot 10^{-11}$
	October	$1,5 \cdot 10^{-11}$
	November	$4,9 \cdot 10^{-11}$
	December	$4,6 \cdot 10^{-11}$
1978	January	$2,1 \cdot 10^{-11}$
	February	$3,2 \cdot 10^{-10}$
	March	$2,6 \cdot 10^{-11}$
	April	$2,0 \cdot 10^{-11}$
	May	$4,0 \cdot 10^{-11}$
	June	$4,8 \cdot 10^{-11}$
	July	$1,8 \cdot 10^{-11}$
	August	$1,2 \cdot 10^{-11}$
	September	$2,0 \cdot 10^{-11}$
	October	$2,9 \cdot 10^{-11}$

Table A 3: Emission of CO_2-bound C-14 in the effluent gas from the Gundremmingen nuclear power plant and the normalized annual emission

A. Annual emission

Year	Mean C-14 concentration (Ci/m^3)	Annual emission (Ci)
1976	$1,9 \cdot 10^{-9}$	3,7
1977	$5,1 \cdot 10^{-10}$	1,0
1978	$5,1 \cdot 10^{-11}$	0,1

B. Normalized annual emission per unit installed electric capacity

Period	Reactor capacity (GW)	Emission (Ci)	Normalized annual emission (Ci/GW)
1976	0,252	3,2	14,7

C. Normalized annual emission from the actual power generated

Period	Power generated (GWh)	Emission (Ci)	Normalized annual emission (Ci/GWa)
01.10.76 to 13.01.77	598	1,02	15,1

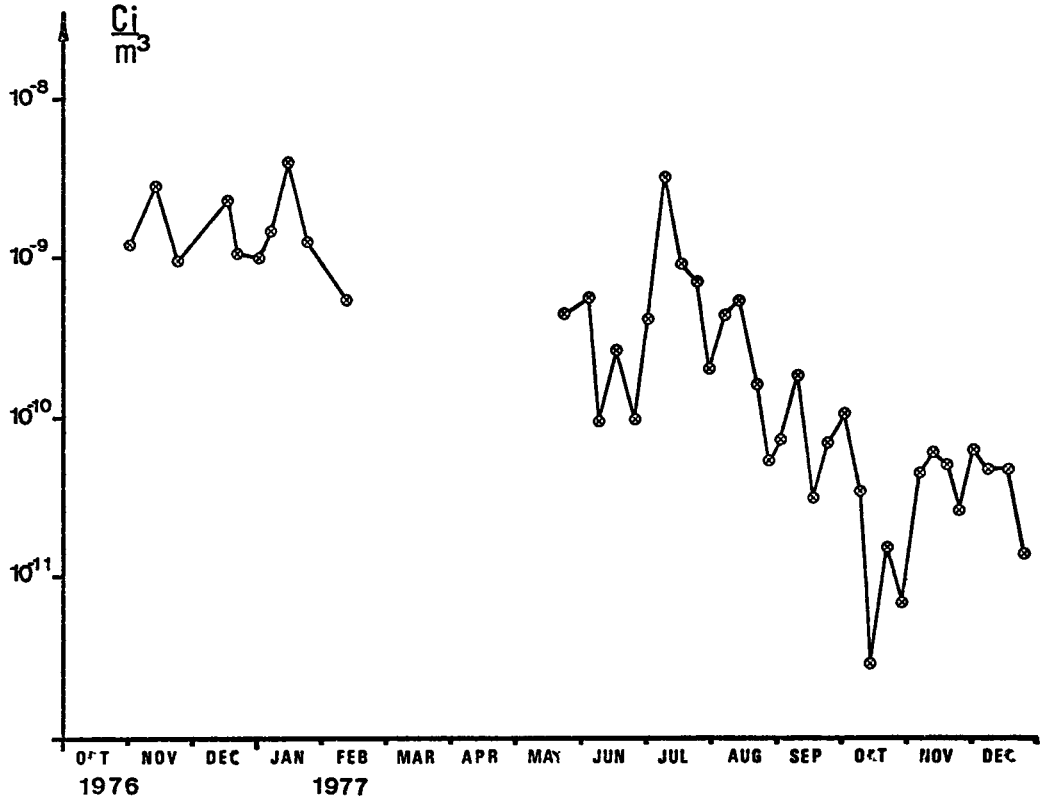

Fig. A1: Time variation of the concentration of CO_2-bound C-14 in the stack effluent gas from the Gundremmingen nuclear plant (reactor closed down from13.01.1977)

Table A4: Concentration of CO_2-bound C-14 in the effluent gas from the Lingen nuclear power plant during normal working of the reactor

Year	Samples taken	C-14 concentration (Ci/m^3)
1976	30.11.	$1,8 \cdot 10^{-9}$
	14.12.	$1,1 \cdot 10^{-9}$
	28.12.	$1,1 \cdot 10^{-9}$

Table A5: Concentration of CO_2-bound C-14 in the effluent gas from the Lingen nuclear power plant after shut-down (monthly mean values)

Year	Samples taken	C-14 concentration (Ci/m^3)
1977	January	$1,7 \cdot 10^{-9}$
	February	$8,5 \cdot 10^{-11}$
	March	$5,3 \cdot 10^{-11}$
	April	$9,3 \cdot 10^{-11}$
	May	$9,3 \cdot 10^{-11}$
	June	$2,8 \cdot 10^{-11}$
	July	$4,7 \cdot 10^{-11}$
	August	$1,5 \cdot 10^{-11}$
	September	$3,8 \cdot 10^{-11}$

Table A6: Emission of CO_2-bound C-14 in the effluent gas from the Lingen nuclear power plant and the normalized annual emission

A. Annual emission

Year	Mean C-14 concentration (Ci/m^3)	Annual emission (Ci)
1976	$1,9,10^{-9}$	1,4
1977	$2,7.10^{-10}$	0,2

B. Normalized annual emission per unit installed electric capacity

Year	Reactor capacity (GW)	Emission (Ci)	Normalized annual emission (Ci/GW)
1976	0,165	1,4	8,5

C. Normalized annual emission from the actual power generated [1)]

Period	Power generated (GWh)	Emission (Ci)	Normalized annual emission (Ci/GWa)
1.11.76 - 5.01.77	240	0,25	9,2

1) For the calculation, the total output or power generated at the Lingen nuclear power plant was taken to be 2/3 from nuclear power and 1/3 from the fossil-fuelled superheater.

(*)

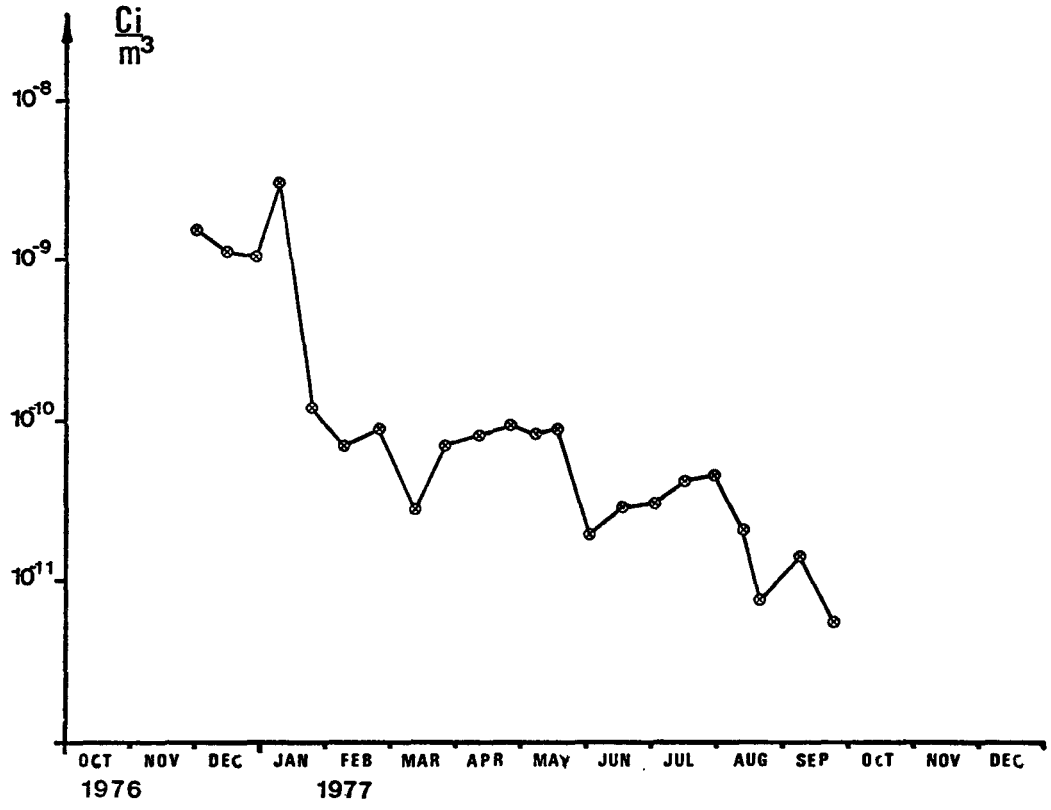

Fig. A2: Time variation of the concentration of CO_2-bound C-14 in the stack effluent gas from the Lingen nuclear plant (reactor closed down 05.01.1977)

Table A7: Concentration of CO_2-bound C-14 in the effluent gas from the Würgassen nuclear power plant (monthly mean values)

Year	Sample taken	C-14 concentration (Ci/m^3)
1977	January	$9{,}0.10^{-10}$
	February	$1{,}0.10^{-9}$
	March	$1{,}3.10^{-9}$
	April	$1{,}3.10^{-9}$
	May	$2{,}4.10^{-9}$
	June	$2{,}0.10^{-9}$
	July	no sample taken
	August	no sample taken
	September	no sample taken
	October	$2{,}6.10^{-10}$
	November	$1{,}5.10^{-9}$
	December	$2{,}8.10^{-9}$
1978	January	$2{,}7.10^{-9}$
	February	$2{,}2.10^{-9}$
	March	$2{,}4.10^{-9}$
	April	$1{,}8.10^{-9}$
	May	$3{,}9.10^{-11}$
	June	$8{,}4.10^{-10}$
	July	$1{,}9.10^{-9}$
	August	no sample taken
	September	$7{,}2.10^{-10}$
	October	$1{,}2.10^{-9}$

Table A8: Emission of CO_2-bound C-14 in the effluent gas from the Würgassen nuclear power plant and the normalized annual emission

A. Annual emission

Year	Mean C-14 concentration (Ci/m^3)	Annual emission (Ci)
1977	$1,5.10^{-9}$	6,2
1978	$1,5.10^{-9}$	6,2

B. Normalized annual emission per unit installed electric capacity

Year	Reactor capacity (GW)	Emission (Ci)	Normalized annual emission (Ci/GW)
1977	0,536	6,2	11,6
1978	0,536	6,2	11,6

C. Normalized annual emission from the actual power generated

Period	Power generated (GWh)	Emission (Ci)	Normalized annual emission (Ci/GWa)
1977	$3,8.10^3$	6,2	14,3
Ist half-year 1978 1)	$1,2.10^3$	3,3	23,8

1) Refitting and change of fuel elements in the first half-year 1978 over about 3 months

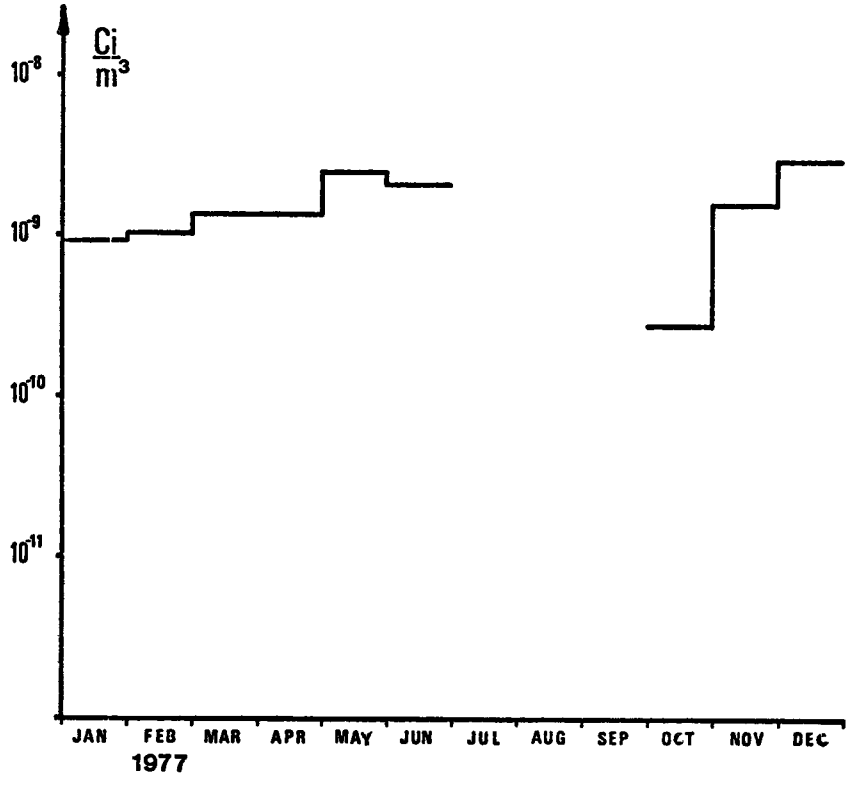

Fig. A3: Time variation of the concentration of CO_2-bound C-14 in the stack effluent gas from Würgassen nuclear plant (monthly total samples)

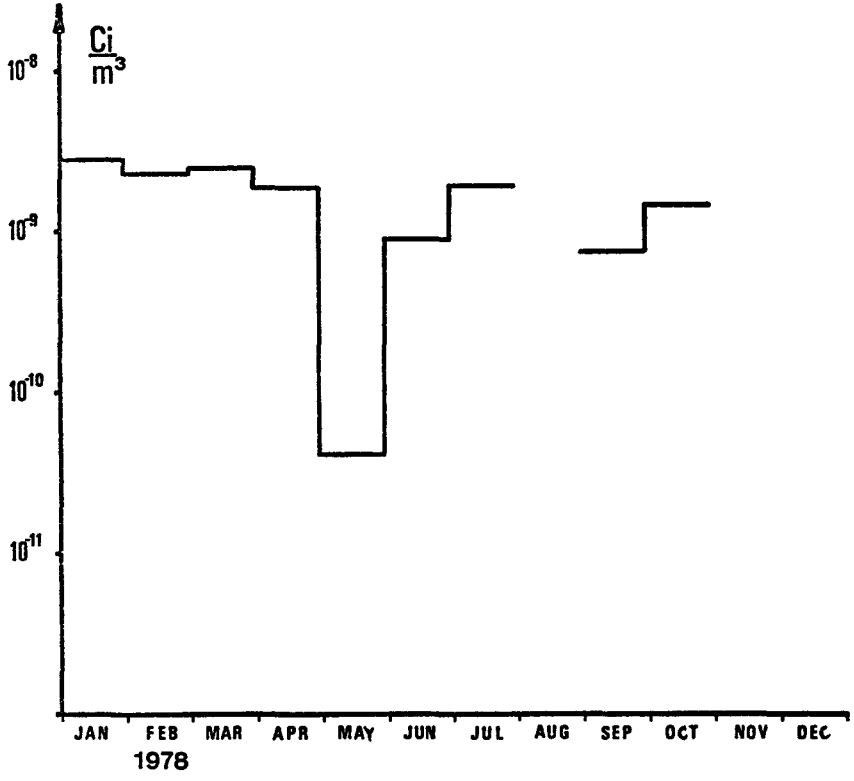

Fig. A4: Time variation of the concentration of CO_2-bound C-14 in the stack effluent gas from the Würgassen nuclear plant (monthly total samples)

Table A9: Concentration of CO_2-bound C-14 in the effluent gas from the Brunsbüttel nuclear power plant (monthly mean values)

Year	Sample taken	C-14 concentration (Ci/m^3)
1977	June	$1,1.10^{-9}$
	July	$7,9.10^{-9}$
	August	$3,0.10^{-10}$
	September	$3,2.10^{-10}$
	October	$1,1.10^{-10}$
	November	$8,2.10^{-12}$
	December	$5,8.10^{-12}$
1978	January	$6,2.10^{-11}$
	February	$2,2.10^{-10}$
	March	$4,3.10^{-9}$
	April	$9,0.10^{-10}$
	May	$1,2.10^{-9}$
	June	$5,7.10^{-9}$
	July	$2,7.10^{-10}$
	August	$2,6.10^{-10}$
	September	$2,6.10^{-10}$
	October	$2,8.10^{-10}$

Table A10: Emission of CO_2-bound C-14 in the effluent gas from the Brunsbüttel nuclear power plant and the normalized annual emission

A. Annual emission

Year	Mean C-14 concentration (Ci/m^3)	Annual emission (Ci)
1977	$1,4.10^{-9}$	4,3
1978	$1,5.10^{-9}$	4,5

B. Normalized annual emission per unit installed electric capacity

Year	Reactor capacity (GW)	Emission (Ci)	Normalized annual emission (Ci/GW)
1977	0,806	4,3	5,3
1978	0,806	4,5	5,6

C. Normalized annual emission from the actual power generated

Period	Power generated (GWh)	Emission (Ci)	Normalized annual emission (Ci/GWa)
1977	$3,5.10^3$	4,3	10,9
First half-year 1978	$2,4.10^3$	3,6	12,9

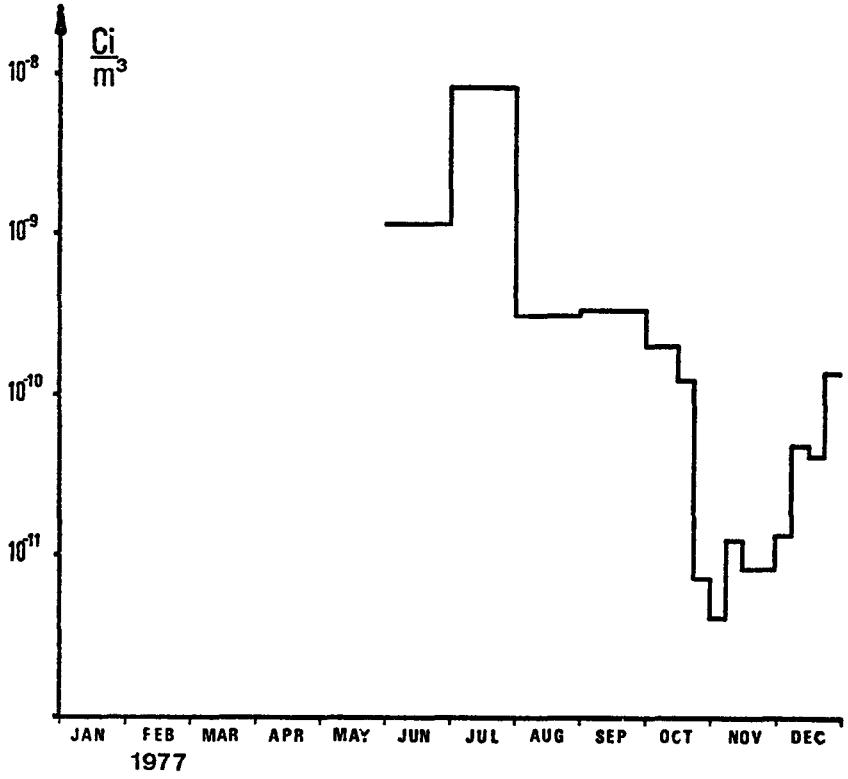

Fig. A5: Time variation of the concentration of CO_2-bound C-14 in the stack effluent gas from the Brunsbüttel nuclear plant (monthly or weekly total samples)

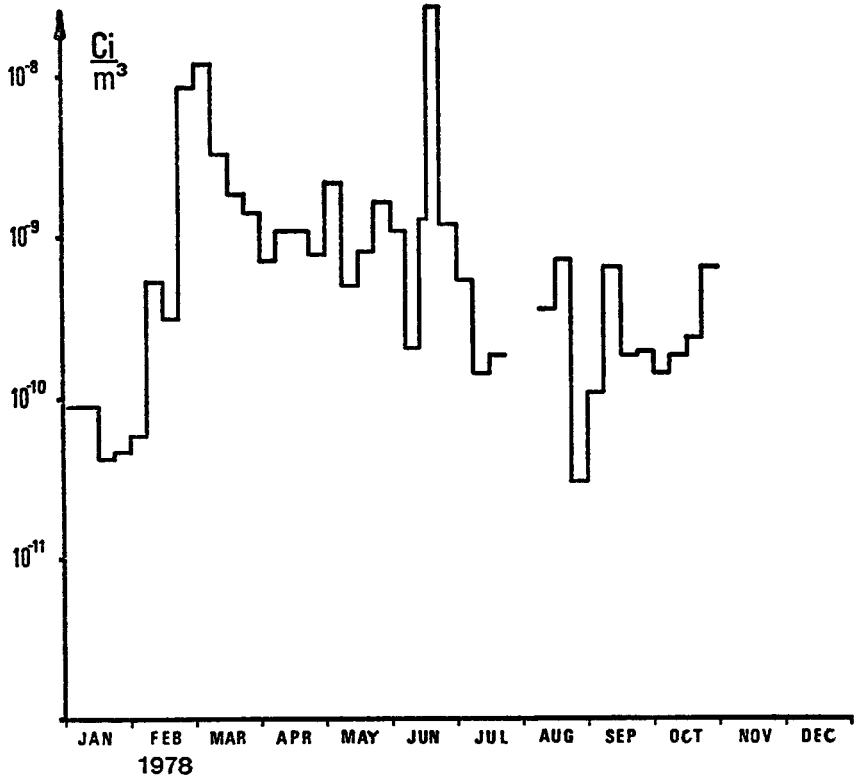

Fig. A6: Time variation of the concentration of CO_2-bound C-14 in the stack effluent gas from the Brunsbüttel nuclear plant (weekly total samples)

Table A11: C-14 concentration in the effluent gas from the Obrigheim nuclear power plant (individual values and percentage CO_2-bound)

Year	Sample taken	C-14 concentration in the effluent gas (Ci/m^3)	Percentage CO_2-bound C-14
1977	03.03.	$1,6.10^{-9}$	14
	29.09.	$8,9.10^{-10}$	30
	01.06.	$3,1.10^{-9}$	42
	30.06.	$3,0.10^{-10}$	40
	27.07.	$2,9.10^{-10}$	24
	06.09.	$1,1.10^{-9}$	12
	06.10.	$3,8.10^{-9}$	3
	09.11.	$1,37.10^{-8}$	5
	15.12.	$1,6.10^{-9}$	26
1978	12.01.	$2,9.10^{-10}$	28
	14.02.	$2,1.10^{-10}$	29
	13.03.	$3,8.10^{-10}$	26
	18.05.	$1,3.10^{-9}$	60
	13.06.	$1,7.10^{-9}$	78
	18.07.	$5,8.10^{-10}$	66
	16.08.	$1,9.10^{-9}$	43
	14.09.	$4,5.10^{-10}$	78
	18.10.	$5,7.10^{-10}$	72

Table A12: C-14 concentration in the effluent gas from the Obrigheim nuclear power plant (continuous sampling)

Year	Period	C-14 concentration in the effluent gas (Ci/m^3)	Percentage CO_2-bound C-14
1978	13.06.-23.06.	$1,1.10^{-9}$	73
	07.09.-19.09.	$6,7.10^{-10}$	81
	19.09.-09.10.	$4,1.10^{-10}$	76
	09.10.-18.10.	$2,6.10^{-9}$	38
	18.10.-31.10.	$5,1.10^{-10}$	78

Table A13: C-14 emission in the effluent gas from the Obrigheim nuclear power plant and the normalized annual emission

A. Annual emission

Year	Mean C-14 concentration	Annual emission of C-14 (Ci)	
	Ci/m^3	total	CO_2-bound
1977	$2,9.10^{-9}$	3,0	0,4
1978	$8,1.10^{-10}$	0,9	0,5

B. Normalized annual emission per unit installed electric capacity

Year	Reactor capacity	Emission	Normalized annual emission
	(GW)	(Ci)	(Ci/GW)
1977	0,345	3,0	8,7
1978	0,345	0,9	2,5

C. Normalized annual emission from the actual power generated

Period	Power generated	Emission	Normalized annual emission
	(GWh)	(Ci)	(Ci/GWa)
1977	$2,3.10^3$	3,0	11,6
First half-year 1978	$1,2.10^3$	0,4	3,0

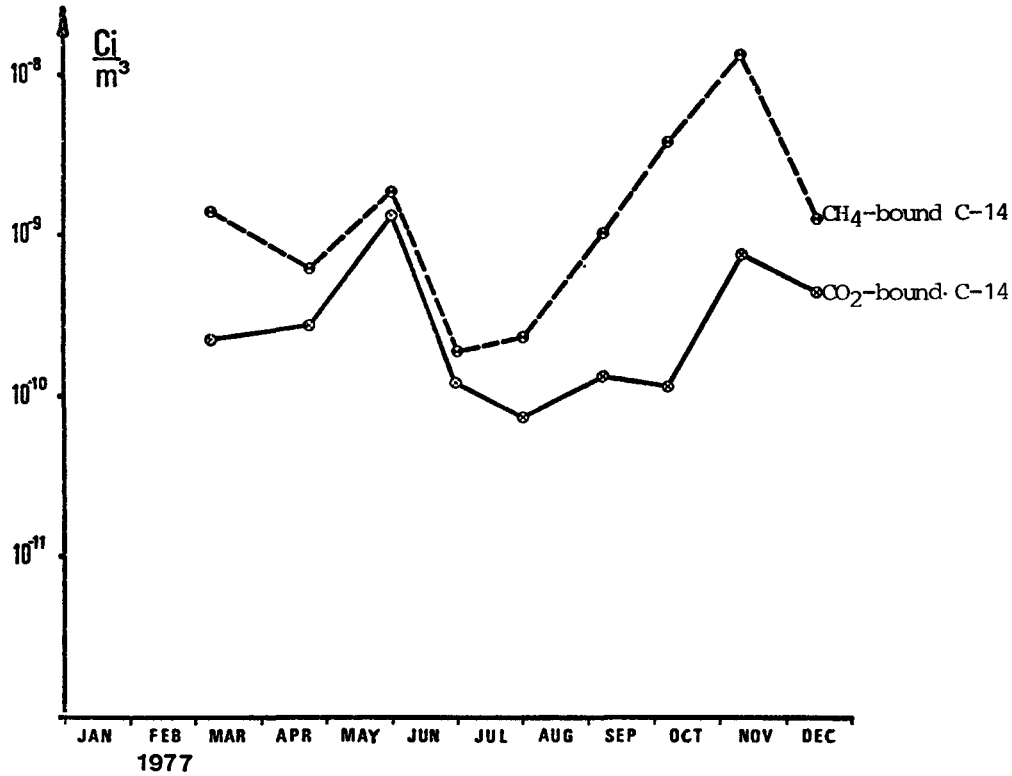

Fig. A7: Time variation of the concentration of C-14 in the stack effluent gas from the Obrigheim nuclear plant (individual measurements)

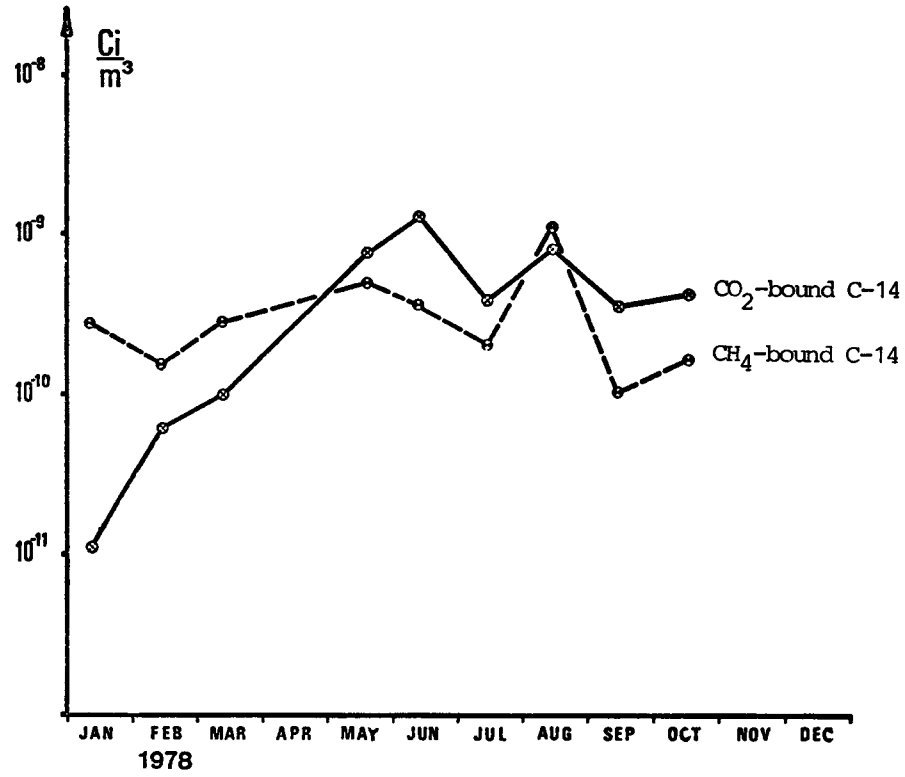

Fig. A8: Time variation of C-14 in the stack effluent gas from the Obrigheim nuclear plant (individual measurements)

Table A14: Concentration of CO_2-bound C-14 in the effluent gas from the Stade nuclear power plant (monthly mean values)

Year	Sample taken	C-14 concentration (Ci/m^3)
1977	April	$1,9 \cdot 10^{-9}$
	May	$1,4 \cdot 10^{-8}$
	June	$1,5 \cdot 10^{-9}$
	July	$4,1 \cdot 10^{-10}$
	August	$3,0 \cdot 10^{-10}$
	September	$2,6 \cdot 10^{-9}$
	October	$2,1 \cdot 10^{-9}$
	November	$5,1 \cdot 10^{-10}$
	December	$4,0 \cdot 10^{-10}$
1978	January	$5,3 \cdot 10^{-10}$
	February	$1,0 \cdot 10^{-9}$
	March	$1,1 \cdot 10^{-9}$
	April	$3,3 \cdot 10^{-9}$
	May	$7,8 \cdot 10^{-10}$
	June	$1,4 \cdot 10^{-9}$
	July	$1,5 \cdot 10^{-9}$
	August	$7,1 \cdot 10^{-10}$
	September	$4,3 \cdot 10^{-10}$
	October	$4,1 \cdot 10^{-10}$

Table A15: C-14 concentration in the effluent gas from the Stade nuclear power plant (complete C-14 separation)

Sample taken	C-14 concentration	CO_2-bound component
01.0 -02.10.78	$8,1.10^{-10}$	53%
02.10-01.11.78	$5,6.10^{-9}$	7%

Table A16: Emission of CO_2-bound C-14 in the effluent gas from the Stade nuclear power plant and the normalized annual emission

A. Annual emission

Year	Mean concentration of CO_2-bound C-14 (Ci/m^3)	Annual emission (only the CO_2-bound component of the C-14) (Ci)
1977	$2,2.10^{-9}$	3,0
1978	$1,1.10^{-9}$	1,5

B. Normalized annual emission per unit installed electric capacity

Year	Reactor capacity (GW)	Emission [1] (Ci)	Normalized annual emission [1] (Ci/GW)
1977	0,662	3,0	4,5
1978	0,662	1,5	2,3

C. Normalized annual emission from the actual power generated

Period	Power generated (GWh)	Emission [1] (Ci)	Normalized annual emission [1] (Ci/GWa)
1977	$5,4.10^3$	3,0	4,8
First half-year 1978	$2,6.10^3$	0,9	3,0

1) Only the CO_2-bound component of the C-14.

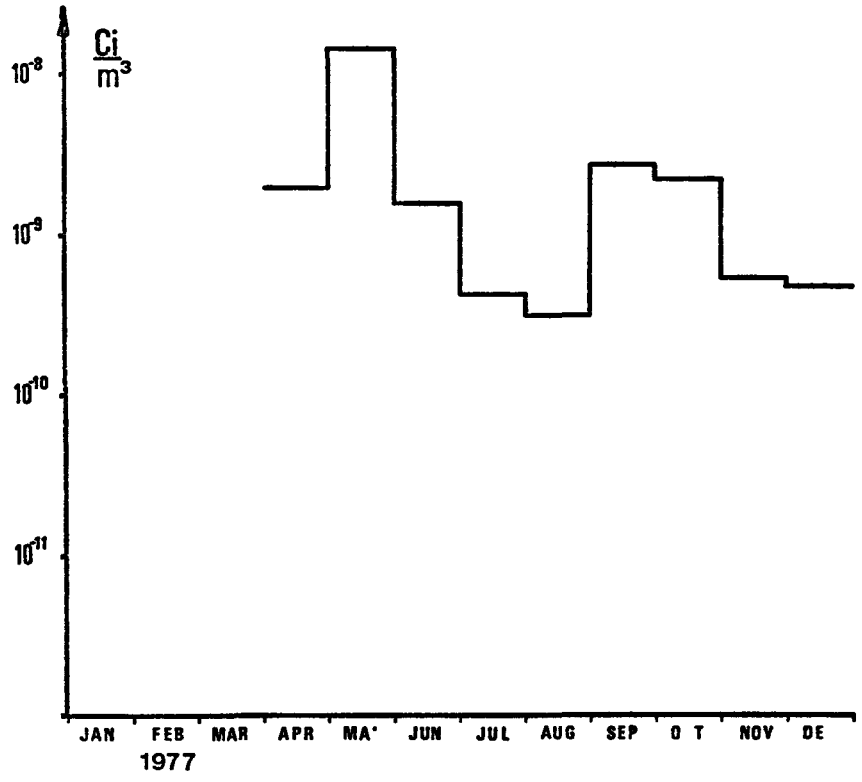

Fig. A9: Time variation of the concentration of CO_2-bound C-14 in the stack effluent gas from the Stade nuclear plant (monthly total samples)

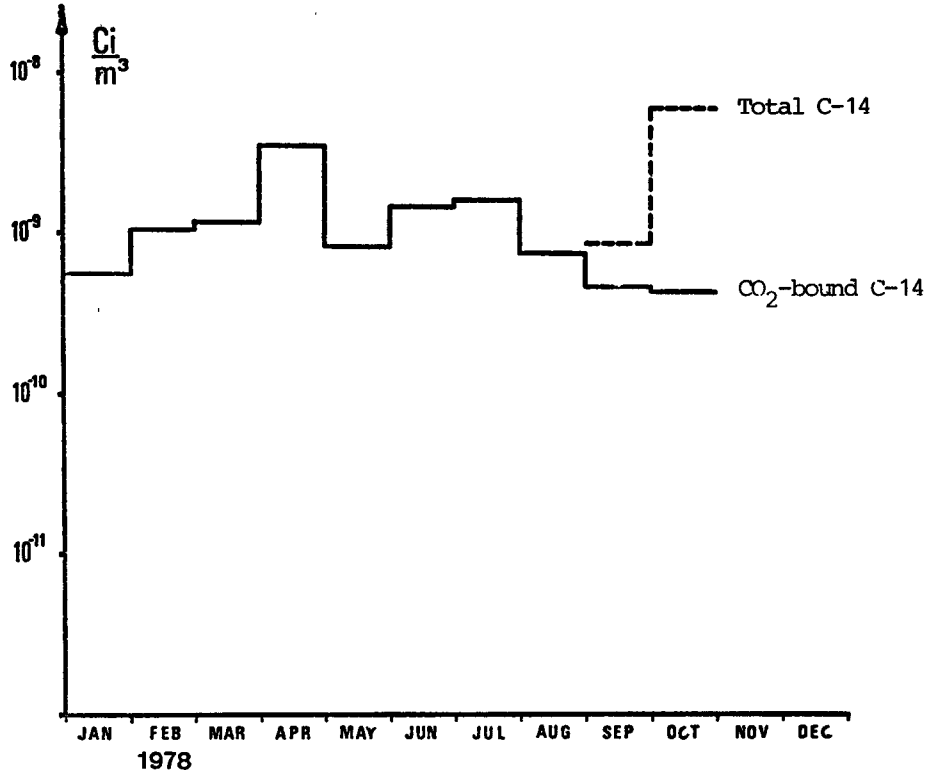

Fig. A10: Time variation of the concentration of CO_2-bound C-14 in the stack effluent gas from the Stade nuclear plant (monthly total samples)

Table A17: C-14 concentration in the effluent gas from the Biblis nuclear power plant, Block A (individual values)

Year	Sample taken	C-14 concentration in the effluent gas in Ci/m^3	Percentage of CO_2-bound C-14
1977	04.03.	$1,8.10^{-9}$	4
	28.04.	$8,0.10^{-10}$	6
	02.06.	$6,8.10^{-10}$	9
	27.07.	$8,3.10^{-10}$	6
	07.09	$6,3.10^{-9}$	3
	05.10.	$2,9.10^{-9}$	49
	09.11.	$9,4.10^{-9}$	4
	14.12.	$4,4.10^{-9}$	7
1978	11.01.	$2,9.10^{-9}$	23
	14.02.	$9,0.10^{-10}$	63
	14.03.	$2,6.10^{-9}$	85
	17.05.	$1,0.10^{-9}$	12
	12.06.	$1,5.10^{-9}$	3
	17.07.	$8,4.10^{-10}$	12
	16.08.	$2,2.10^{-9}$	6
	15.09.	$8,5.10^{-10}$	5
	18.10.	$3,4.10^{-10}$	13

Table 18: Emission of C-14 in the effluent gas from the Biblis nuclear power station, Block A, and the normalized annual emission

A. Annual emission

Year	Mean C-14 concentration (Ci/m^3)	Annual emission of C-14 (Ci)	
		total	Co_2-bound
1977	$3,4.10^{-9}$	4,8	0,4
1978	$1,5.10^{-9}$	2,1	0,6

B. Normalized annual emission per unit installed electric capacity

Year	Reactor capacity (GW)	Emission (Ci)	Normalized annual emission (Ci/GW)
1977	1,204	4,8	4,0
1978	1,204	2,1	1,3

C. Normalized annual emission from the actual power generated

Period	Power generated (GWh)	Emission (Ci)	Normalized annual emission (Ci/GWa)
1977	$6,6.10^3$	4,8	6,4
First half-year 1978	$5,2.10^3$	1,2	2,0

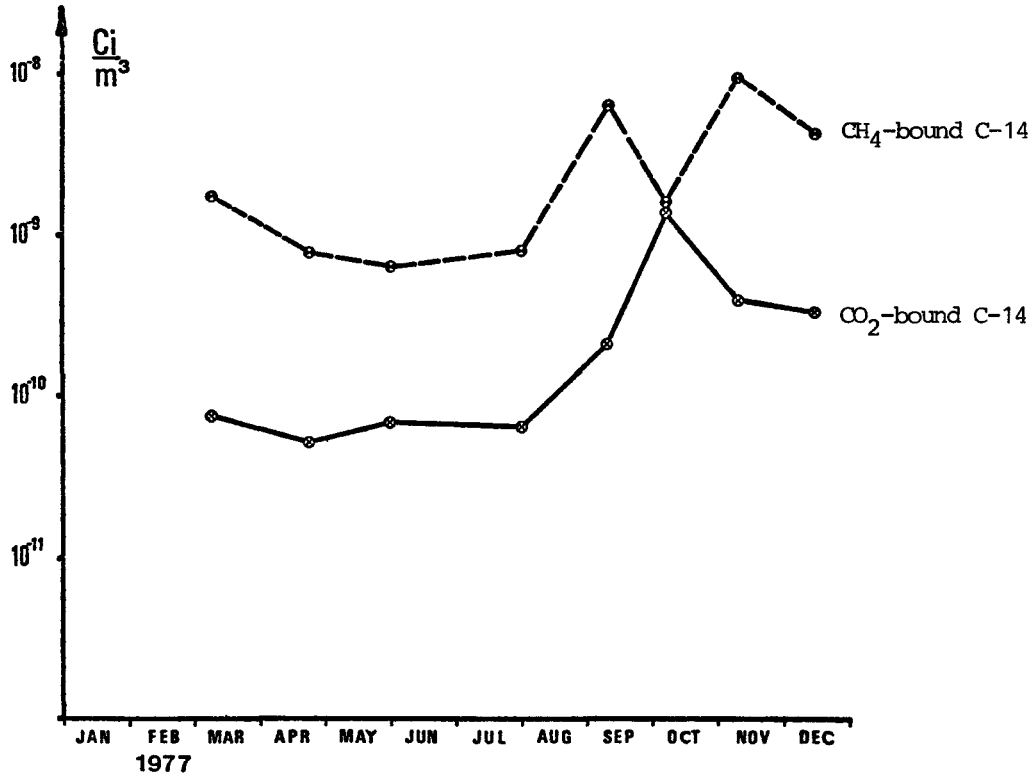

Fig. A11: Time variation of the concentration of C-14 in the stack effluent gas from the Biblis nuclear power plant, Block A (individual measurements)

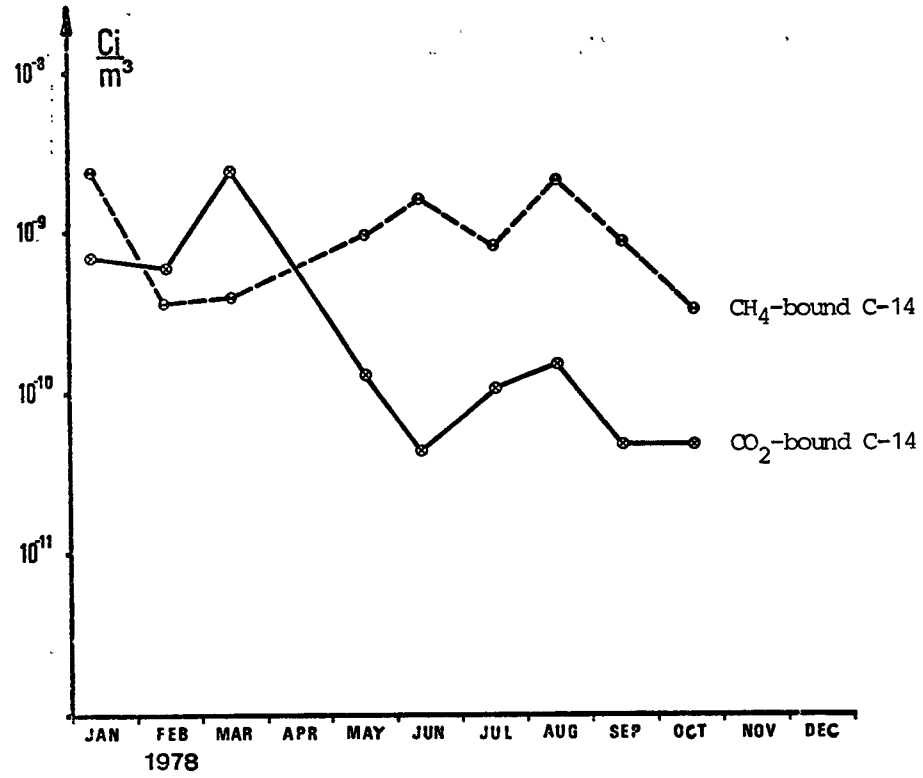

Fig. A12: Time variation of the concentration of C-14 in the stack effluent gas from the Biblis nuclear power plant, Block A (individual measurements)

Table A19: Concentration of CO_2-bound C-14 in the effluent gas from the Biblis nuclear power plant, Block B (monthly values)

Year	Period	Concentration of CO_2-bound C-14 in the effluent gas in Ci/m^3
1977	May	$6,8 \cdot 10^{-11}$
	June	$1,7 \cdot 10^{-10}$
	July	$2,1 \cdot 10^{-10}$
	August	$0,5 \cdot 10^{-12}$
	September	$3,3 \cdot 10^{-11}$
	October	$8,0 \cdot 10^{-11}$
	November	$1,2 \cdot 10^{-10}$
	December	$1,7 \cdot 10^{-11}$
1978	January	$3,2 \cdot 10^{-10}$
	February	$1,7 \cdot 10^{-10}$
	March	$2,6 \cdot 10^{-10}$
	April	$2,6 \cdot 10^{-10}$
	May	$1,4 \cdot 10^{-10}$
	June	$3,1 \cdot 10^{-10}$
	July	$1,8 \cdot 10^{-10}$
	August	$6,4 \cdot 10^{-10}$
	September	$4,2 \cdot 10^{-10}$

Table A20: C-14 concentration in the effluent gas from the Biblis nuclear power plant, Block B (individual measurements)

Year	Sample taken	C-14 concentration in the effluent gas in Ci/m^3	Percentage of CO_2-bound C-14
1978	11.01.	$7,9 \cdot 10^{-9}$	3
	14.02.	$4,2 \cdot 10^{-10}$	62
	14.03.	$4,4 \cdot 10^{-10}$	52
	17.05.	$4,5 \cdot 10^{-9}$	6
	12.06.	$3,4 \cdot 10^{-9}$	7
	12.07.	$4,6 \cdot 10^{-10}$	37
	16.08.	$9,1 \cdot 10^{-10}$	8
	15.09.	$9,0 \cdot 10^{-10}$	15
	18.10.	$2,5 \cdot 10^{-9}$	11

Table 21: Emission of C-14 in the effluent gas from the Biblis nuclear power plant, Block B, and the normalized annual emission

A. Annual emission

Year	Mean C-14 concentration in the effluent gas (Ci/m^3)	Annual emission of C-14 (Ci)	
		total	CO_2-bound
1977	$3,5 \cdot 10^{-9}$	4,9	0,12
1978	$3,4 \cdot 10^{-9}$	4,8	0,42

B. Normalized annual emission per unit installed electric capacity

Year	Reactor capacity (GW)	Emission (Ci)	Normalized annual emission (Ci/GW)
1977	1,300	4,9	3,8
1978	1,300	4,8	3,7

C. Normalized annual emission from the actual power generated

Period	Power generated (GWh)	Emission (Ci)	Normalized annual emission (Ci/GWa)
1977	$9,5 \cdot 10^3$	4,5	5,0
First half-year 1978	$2,4 \cdot 10^3$	2,0	7,2

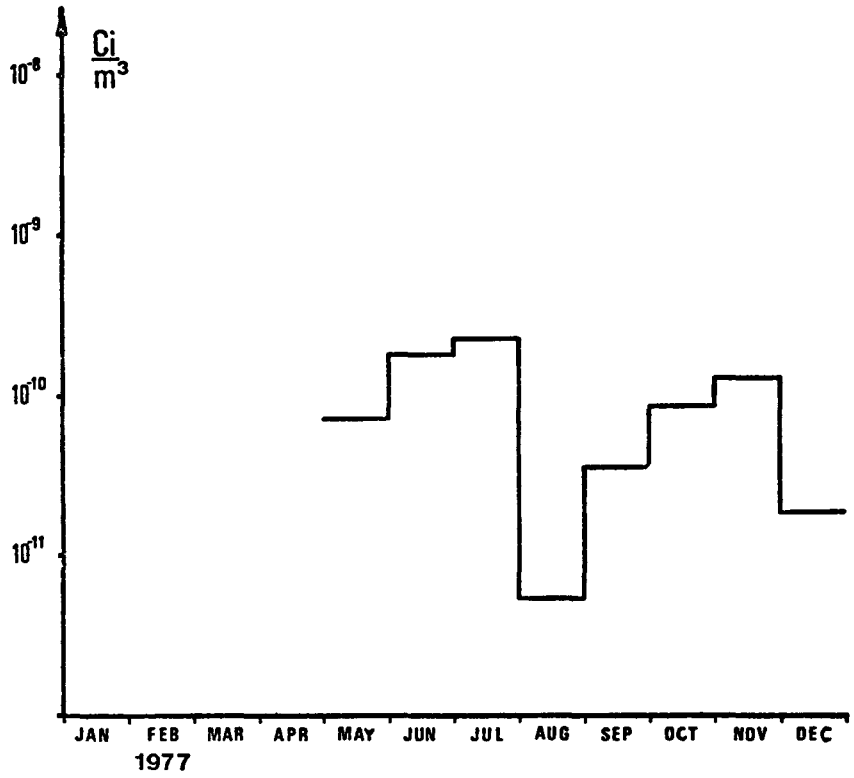

Fig. A13: Time variation of the concentration of CO_2-bound C-14 in the stack effluent gas from the Biblis nuclear power plant, Block B (monthly total samples)

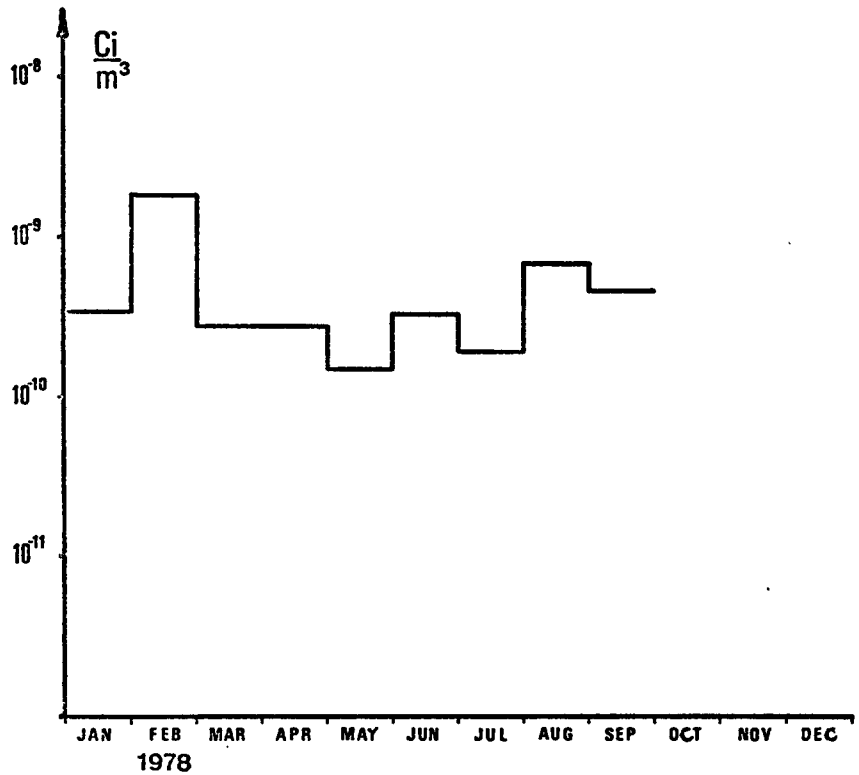

Fig. A14: Time variation of the concentration of CO_2-bound C-14 in the stack effluent gas from the Biblis nuclear power plant, Block B
(monthly total samples)

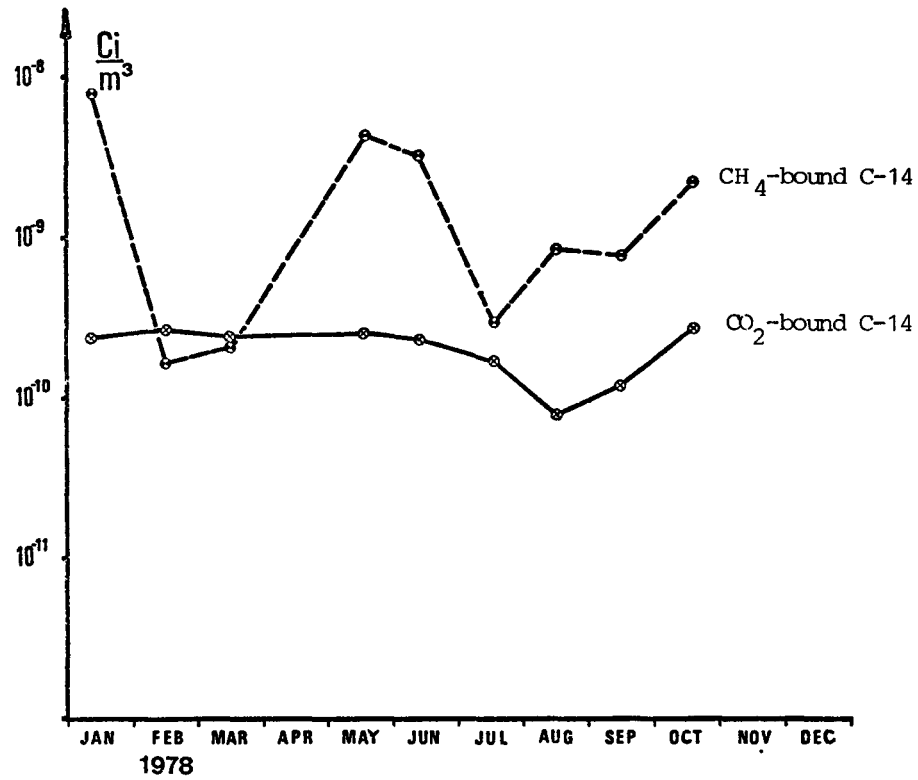

Fig. A15: Time variation of the concentration of C-14 in the stack effluent gas from the Biblis nuclear power plant, Block B (individual measurements)

Table A22: Concentration of CO_2-bound C-14 in the stack gas of the Neckarwestheim nuclear power plant (continuous separation)

Year	Period	C-14 concentration in the effluent gas in Ci/m^3
1977	August	$3,2 \cdot 10^{-10}$
	September	$3,8 \cdot 10^{-10}$
	October	$1,9 \cdot 10^{-10}$
	November	$1,5 \cdot 10^{-10}$
	December	$5,8 \cdot 10^{-11}$
1978	January	$3,7 \cdot 10^{-11}$
	February	$2,7 \cdot 10^{-11}$
	March	$2,8 \cdot 10^{-11}$
	April	$3,1 \cdot 10^{-11}$
	May	$1,8 \cdot 10^{-10}$
	June	$7,9 \cdot 10^{-11}$
	July	$1,3 \cdot 10^{-10}$
	August	$6,8 \cdot 10^{-11}$
	September	$4,3 \cdot 10^{-11}$
	October	$4,6 \cdot 10^{-10}$

Table A23: C-14 concentration in the effluent gas from the Neckarwestheim nuclear power plant (individual measurements)

Year	Sample taken	C-14 concentration in the effluent gas in Ci/m^3	Percentage of CO_2-bound C-14
1977	03.03	$2,8 \cdot 10^{-10}$	11
	01.06	$1,9 \cdot 10^{-9}$	4
	30.06	$5,9 \cdot 10^{-10}$	7
	27.07.	$1,4 \cdot 10^{-10}$	17
	06.09.	$4,8 \cdot 10^{-9}$	1
	06.10.	$4,5 \cdot 10^{-9}$	3
	09.11.	$5,0 \cdot 10^{-9}$	1
	15.12.	$1,1 \cdot 10^{-9}$	9
1978	12.01.	$4,0 \cdot 10^{-9}$	3
	15.02.	$2,8 \cdot 10^{-10}$	21
	13.03.	$4,1 \cdot 10^{-9}$	1
	18.05.	$1,2 \cdot 10^{-9}$	5
	13.06.	$3,2 \cdot 10^{-9}$	2
	18.07.	$2,3 \cdot 10^{-9}$	4
	14.08.	$5,6 \cdot 10^{-10}$	14
	14.09.	$2,2 \cdot 10^{-10}$	89
	18.10.	$2,8 \cdot 10^{-10}$	25

Table A 24: C-14 emission in the effluent gas from the Neckarwestheim nuclear power plant and the normalized annual emission

A. Annual emission

Year	Mean C-14 concentration (Ci/m^3)	Annual emission of C-14 (Ci)	
		total	CO_2-bound
1977	$2,3 \cdot 10^{-9}$	4,0	0,11
1978	$2,2 \cdot 10^{-9}$	3,9	0,16

B. Normalized annual emission per unit installed electric capacity

Year	Reactor capacity	Emission	Normalized annual emission
	(GW)	(Ci)	(Ci/GW)
1977	0,855	4,0	4,7
1978	0,855	3,9	4,6

C. Normalized annual emission from the actual power generated

Period	Power generated	Emission	Normalized annual emission
	(GWh)	(Ci)	(Ci/GWa)
1977	$5,3.10^3$	4,0	6,6
First half-year 1978	$3,1.10^3$	2,2	6,3

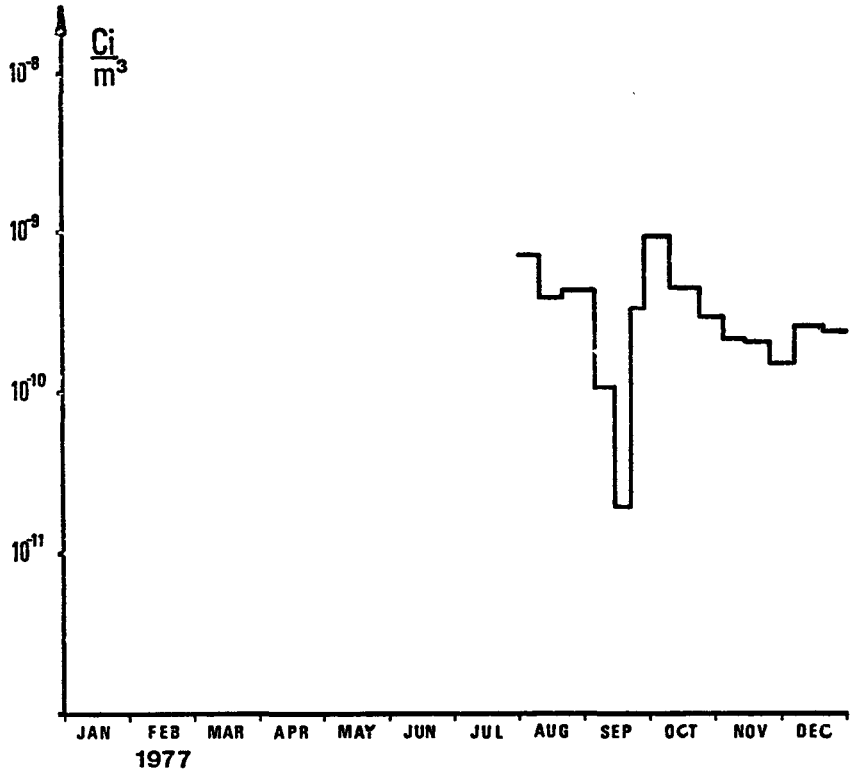

Fig. A16: Time variation of the concentration of CO_2-bound C-14 in the stack effluent gas from the Neckarwestheim nuclear plant (weekly total samples)

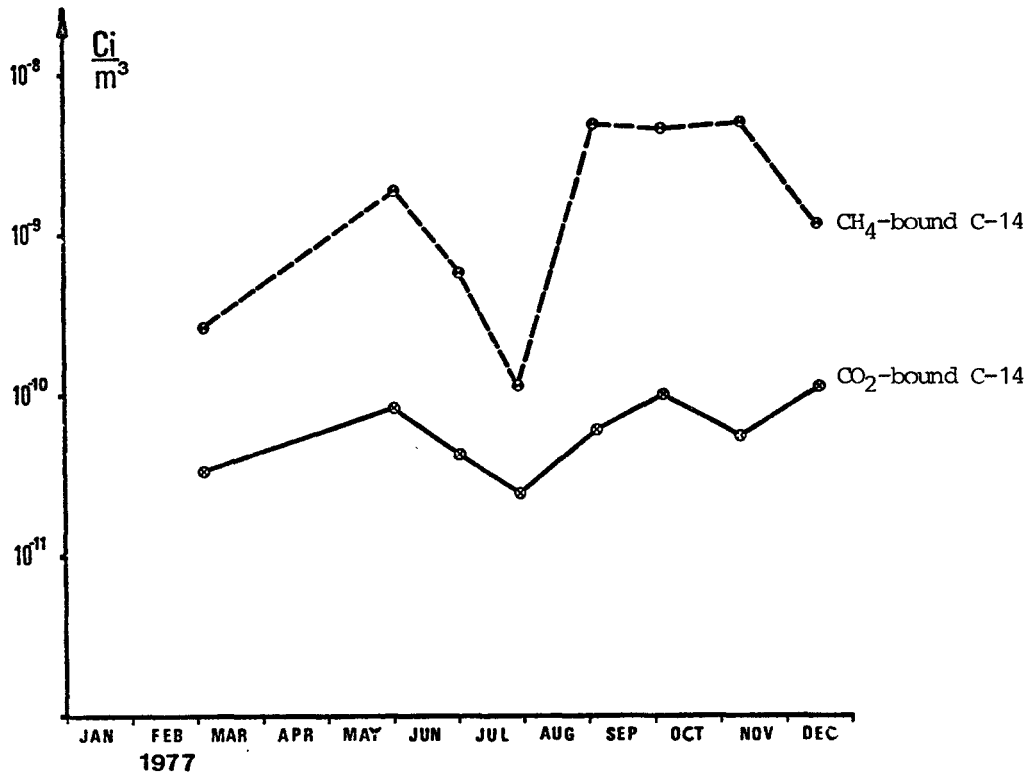

Fig. A17: Time variation of the concentration of C-14 in the stack effluent gas from the Neckarwestheim nuclear plant (individual measurements)

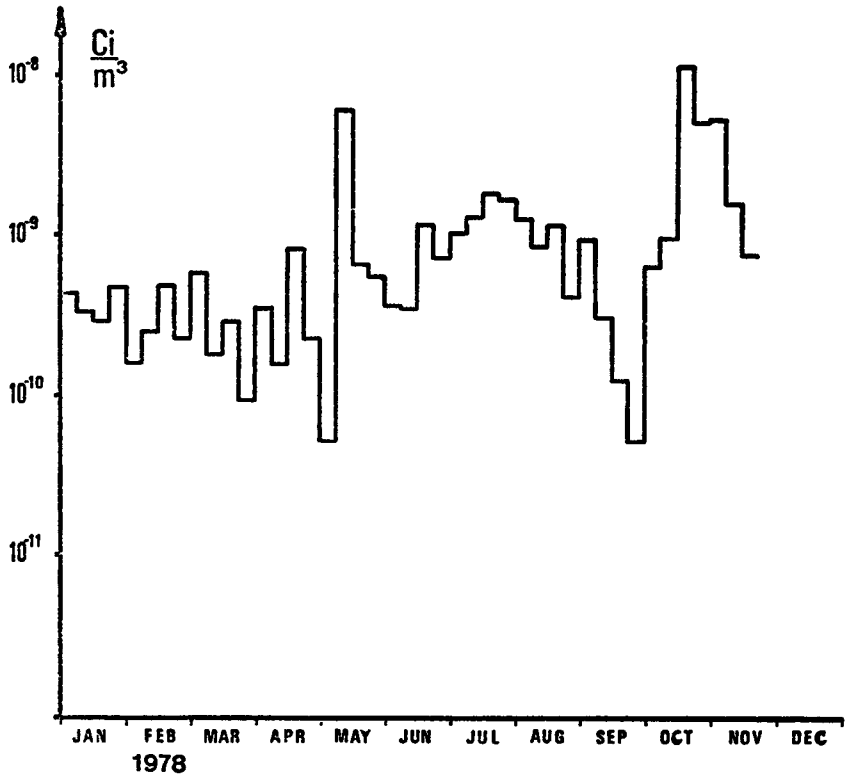

Fig. A18: Time variation of the concentration of CO_2-bound C-14 in the stack effluent gas from the Neckarwestheim nuclear plant (weekly total samples)

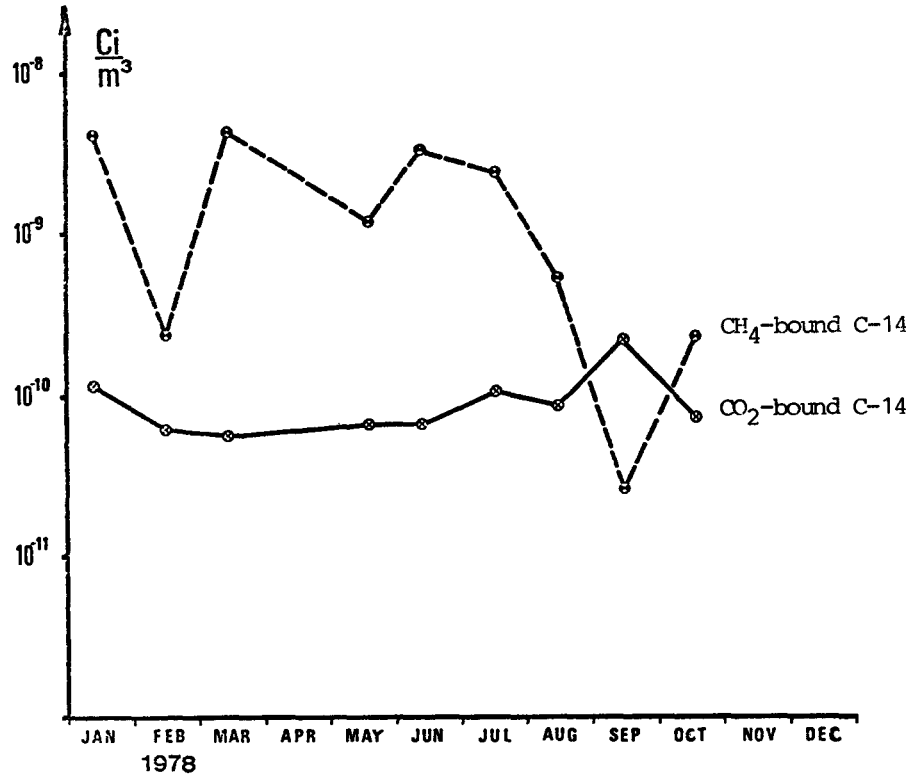

Fig. A19: Time variation of the concentration of C-14 in the stack effluent gas from the Neckarwestheim nuclear plant (individual measurements)

Table A25: Air throughput from the separate parts of the Brunsbüttel nuclear power plant to the stack.

		m^3/h
Reactor building and auxiliary building	ca.	170 000
a) Ventilation air from reactor and auxiliary building	ca.	150 000
b) Scavenging air from the reactor building	ca.	17 000
c) Interspace extract	ca.	30
d) Pump seal exhaust	ca.	200
Turbine hall		140 000
Control rooms [1]		40 000
Off-gas from the absorption system		17
Stack effluent gas	ca.	350 000

1) No C-14 measurements were made here

Table A26: C-14 measurements in the separate lines to the discharge stack at Brunsbüttel nuclear power plant for the period 14-16 June 1978

Channel	C-14 concentration (Ci/m^3)	Rate of emission of C-14 (Ci/h)	Percentage of total emission %
Turbine hall effluent gas	$7{,}9.10^{-10}$	$1{,}2.10^{-4}$	29
Reactor building and auxiliary building effluent	$4{,}3.10^{-12}$	$0{,}06.10^{-4}$	2
Reactor building ventilation air	$2{,}2.10^{-9}$	$0{,}4.10^{-4}$	9
Off-gas system	$9{,}9.10^{-6}$	$1{,}2.10^{-4}$	42
Others [1]	--	$0{,}7.10^{-4}$	18
Stack effluent gas	$1{,}2.10^{-9}$	$3{,}98.10^{-4}$	100

1) No measurement in air from the control rooms, interspace and pump seal extracts

Table A27: Air throughput from the separate parts of the Neckarwestheim nuclear power plant to the stack

	m^3/h
Reactor building:	
Locked off equipment rooms	70 000
Operating and equipment rooms	1 000
Auxiliary equipment buildings:	
filtered	20 000
unfiltered	80 000
Off-gas	0-5
Stack	171 000

Table 28: C-14 measurements in the separate lines to the discharge stack at the Neckarwestheim nuclear power plant for the period 14-16 August 1978

Channel	C-14 concentration	Rate of emission of C-14	Percentage of total emission
	(Ci/m^3)	(Ci/h)	%
Reactor building:			
Locked off equipment rooms	$1,3.10^{-10}$	$9,1.10^{-6}$	8
Operating and equipment rooms	$8,8.10^{-6}$	$8,8.10^{-5}$	76
Auxiliary equipment buildings:			
filtered	$5,2.10^{-10}$	$1,0.10^{-5}$	9
unfiltered	$1,1.10^{-10}$	$8,8.10^{-6}$	7
Stack effluent gas	$5,6.10^{-10}$	$9,6.10^{-5}$	100

Table A29: Concentration of C-14 in the effluent gas from the Neckarwestheim nuclear power plant

Date	Concentration (Ci/m^3)	Percentage of CO_2-bound C-14 %
14.08.1978	$7,9.10^{-4}$	0,6
15.08.1978	$7,9.10^{-4}$	0,7
16.08.1978	$8,1.10^{-4}$	0,7

Table A30: Concentration of C-14 in the effluent gas from the reprocessing plant, Karlsruhe

Sample number	Period Date	Time	Concentration of C-14 $^{14}CO_2$ (Ci/m^3)	$^{14}CO, ^{14}CH_4$ (Ci/m^3)
1	19.04.	10.00 - 11.00	$4,04 - 10^{-8}$	$3,8 \cdot 10^{-11}$
2	19.04.	17.00 - 19.00	$8,10 \cdot 10^{-8}$	$1,04 \cdot 10^{-10}$
3	19.04.	19.00 - 20.00	$1,49 \cdot 10^{-6}$	$3,47 \cdot 10^{-9}$
4	19.04.	20.00 - 21.00	$1,69 \cdot 10^{-5}$	$2,75 \cdot 10^{-8}$
5	19.04.	21.00 - 22.00	$1,30 \cdot 10^{-5}$	$1,4 \cdot 10^{-8}$
6	19.04.	22.00 - 23.00	$3,53 \cdot 10^{-6}$	$1,34 \cdot 10^{-9}$
7	19.04.	23.00 - 24.00	$1,38 \cdot 10^{-6}$	$4,92 \cdot 10^{-10}$
8	20.04.	0.00 - 1.00	$1,17 \cdot 10^{-6}$	$7,2 \cdot 10^{-11}$
9	20.04.	1.00 - 2.00	$6,99 \cdot 10^{-7}$	$3,4 \cdot 10^{-11}$
10	20.04.	2.00 - 3.00	$3,36 \cdot 10^{-7}$	$4,0 \cdot 10^{-11}$
11	20.04.	3.00 - 4.00	$3,61 \cdot 10^{-7}$	$3,8 \cdot 10^{-11}$
12	20.04.	4.00 - 5.00	$1,46 \cdot 10^{-7}$	$3,5 \cdot 10^{-11}$
13	20.04.	5.00 - 6.00	$4,70 \cdot 10^{-8}$	$4,2 \cdot 10^{-11}$
14	20.04.	6.00 - 8.00	$8,37 \cdot 10^{-8}$	$3,4 \cdot 10^{-11}$
15	20.04.	8.00 - 10.00	$5,92 \cdot 10^{-8}$	$4,4 \cdot 10^{-11}$
16	20.04.	10.00 - 12.00	$4,01 \cdot 10^{-8}$	$3,2 \cdot 10^{-11}$

Table A31: Emission of C-14 in the effluent gas from the reprocessing plant, Karlsruhe

Sample number	Period Date	Time	Emission of C-14 in the effluent gas $^{14}CO_2$	^{14}CO, $^{14}CH_4$
			µCi	µCi
1	19.04.	10.00 - 11.00	20,2	0,019
2	19.04.	17.00 - 19.00	81,0	0,104
3	19.04.	19.00 - 20.00	745	1,735
4	19.04.	20.00 - 21.00	8450	13,7
5	19.04.	21.00 - 22.00	6500	7,0
6	19.04.	22.00 - 23.00	1765	0,67
7	19.04.	23.00 - 24.00	690	0,246
8	20.04.	0.00 - 1.00	585	0,036
9	20.04.	1.00 - 2.00	349,5	0,017
10	20.04.	2.00 - 3.00	168	0,020
11	20.04.	3.00 - 4.00	180,5	0,019
12	20.04.	4.00 - 5.00	73	0,018
13	20.04.	5.00 - 6.00	23,5	0,021
14	20.04.	6.00 - 8.00	83,7	0,034
15	20.04.	8.00 - 10.00	59,2	0,044
16	20.04.	10.00 - 12.00	40,1	0,032
2-16	19.04. 17.00 20.04. 12.00		19,8 mCi	23,7 µCi

Table A32: Percentage of ^{14}CO and $^{14}CH_4$ in the total emission from the reprocessing plant, Karlsruhe

Sample number	Period Date	Time	^{14}CO, $^{14}CH_4$ %
1	19.04.	10.00 - 11.00	0,09
2	19.04.	17.00 - 19.00	0,13
3	19.04.	19.00 - 20.00	0,23
4	19.04.	20.00 - 21.00	0,16
5	19.04.	21.00 - 22.00	0,11
6	19.04.	22.00 - 23.00	0,04
7	19.04.	23.00 - 24.00	0,04
8	20.04.	0.00 - 1.00	0,006
9	20.04.	1.00 - 2.00	0,01
10	20.04.	2.00 - 3.00	0,01
11	20.04.	3.00 - 4.00	0,01
12	20.04.	4.00 - 5.00	0,02
13	20.04.	5.00 - 6.00	0,09
14	20.04.	6.00 - 8.00	0,04
15	20.04.	8.00 - 10.00	0,07
16	20.04.	10.00 - 12.00	0,02

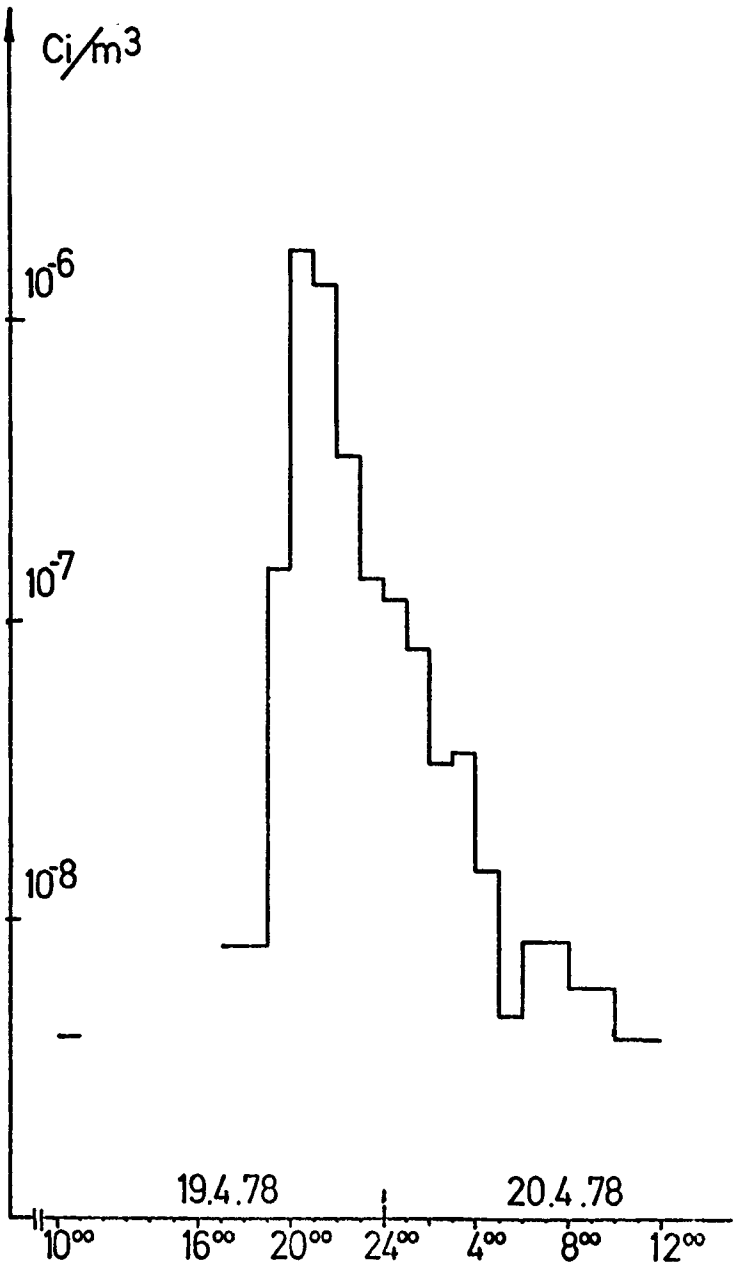

Fig. A 20: Time variation of the concentration of CO_2-bound C-14 in the stack effluent at WAK during a dissolution cycle

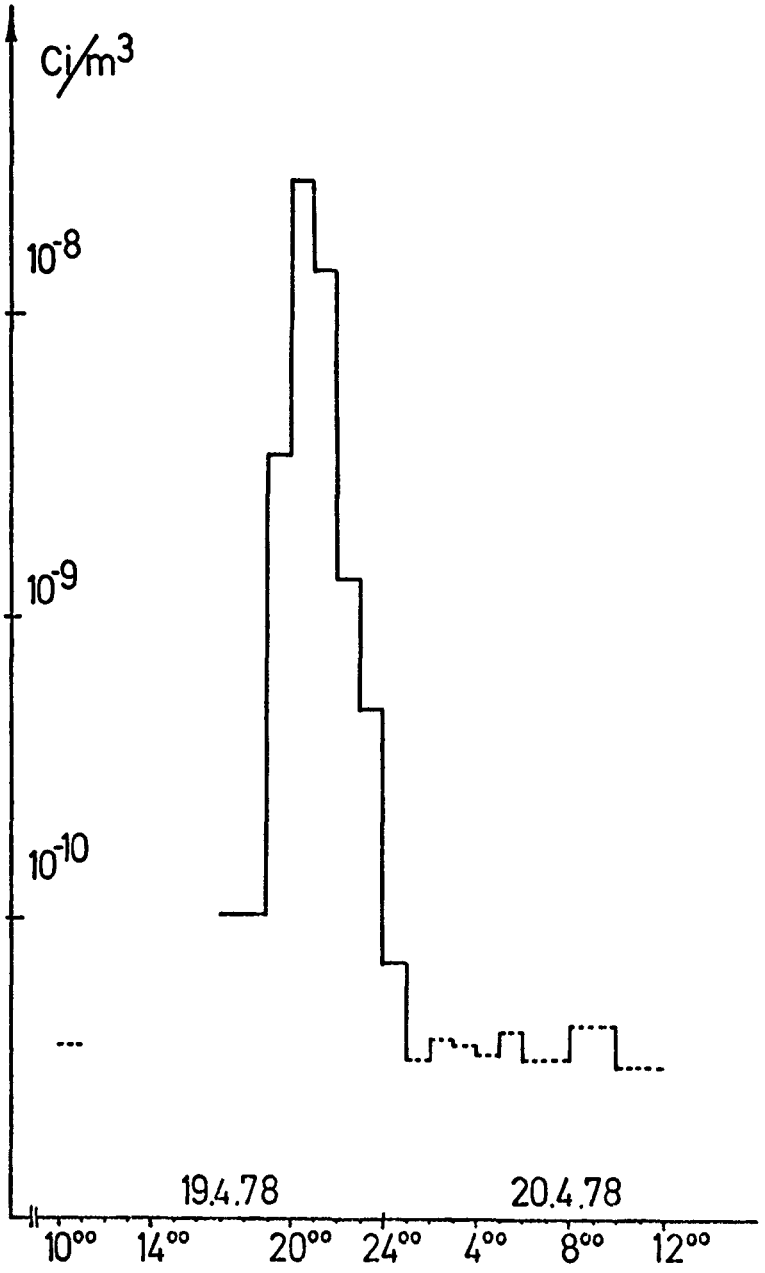

Fig. A21: Time variation of the concentration of CO-bound and hydrocarbon-bound C-14 in the stack effluent at WAK during a dissolution cycle