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Source terms, isolation and radiological consequences of carbon-14 waste in the Swedish SFR repository

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SOURCE TERMS, ISOLATION AND RADIOLOGICAL CONSEQUENCES OF CARBON-14 WASTE IN THE SWEDISH SFR REPOSITORY

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Abstract

The source term, isolation capacity, and long-term radiological exposure of ^{14}C from the Swedish underground repository for low and intermediate level waste (SFR) is assessed. The prospective amount of ^{14}C in the repository is assumed to be 5 TBq. Spent ion exchange resins will be the dominant source of ^{14}C. The pore water in the concrete repository is expected to maintain a pH of >10.5 for a period of at least 10^6 y. The cement matrix of the repository will retain most of the $^{14}CO_3^{2-}$ initially present. Bacterial production of CO_2 and CH_4 from degradation of ion-exchange resins and bitumen may contribute to ^{14}C release to the biosphere. However, CH_4 contributes only to a small extent to the overall carbon loss from freshwater ecosystems. The individual doses to local and regional individuals peaked with $5 \cdot 10^{-3}$ and $8 \cdot 10^{-4}$ µSv y^{-1} respectively at about $2.4 \cdot 10^4$ years. A total leakage of 8.4 GBq of ^{14}C from the repository will cause a total collective dose commitment of 1.1 manSv or 130 manSv TBq^{-1}.

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General Introduction

The Swedish repository for Low and Intermediate Level Radioactive Waste (SFR) is situated at Forsmark close to the Baltic Sea coast, approximately 50 m below the rock surface. Studies on the consequences of possible leakage from the SFR have identified C-14 as one of the most important nuclides in the inventory (Bergström and Puigdomenech, 1987; Wiborgh et al.,1987b) in agreement with other studies (Pigford, 1984; Malbrain and Lester, 1987; Oversby, 1987; Burkholder et al.,1976; Vieno et al.,1987; Häggblom and Kjellbert, 1979). C-14 is of special concern because of its long half-life (5730 y) and because it is assimilated by plants from the atmospheric carbon dioxide pool through photosynthesis, being thus incorporated in the human food chain. The relative importance of C-14 as a radiation source from both nuclear explosions and nuclear power production is documented by UNSCEAR (1982).

In the SFR repository, spent ion exchange resins will be the predominant source of C-14. Due to the difficulties to detect C-14 properly, efforts have been made to establish scaling factors between C-14 and other radionuclides more easily recorded (EPRI, 1980; Cline et al, 1985). However, the lack of similarity in behaviour with other radionuclides, together with large variations in the C-14 concentrations in the reactor process, makes the quantification of C-14 production and inventory uncertain. In the first part of this report (pp 1:1 - 1:19), different ways of asessing the C-14 content in spent ion exchange resins are compared, and the potential source of C-14 in the SFR repository is estimated.

The changes expected to take place in the repository are due to interactions between different kinds of materials and between repository components and the groundwater. Radiolysis, microbial degradation and corrosion of metals will contribute to gas formation, while other chemical reactions will degrade ion exchange resins, precipitate carbonates etc. Furthermore, concrete and bentonite degradation will affect the hydraulic properties of the repository. In the second part of this report (pp 2:1 - 2:30), a survey is made of the processes occurring in a geological repository, with special emphasis on the concrete silo in the SFR repository.

The individual and collective doses resulting from the long-term release of C-14 from the SFR installation is assessed in the third part (pp 3:1 - 3:78). As discussed by Bergström and Puigdomenech (1987), the highest calculated radiological consequences from any radionuclide leakage correspond to a scenario where the underground repository remains intact for 2500 years. During that time the rising of the Scandinavian peninsula will lift the repository site above the Baltic sea surface, and the radionuclides might be discharged directly either to a well, a small lake, or to agricultural land. The chosen starting time for radionuclide release - 2500 years after repository closure - is the earliest time when a lake might form within that part of the area.

Source terms of Carbon-14 in SFR-1 waste

1 Introduction

The waste-type in the SFR that dominates the radionuclide inventory is spent ion exchange resins from the nuclear power plants in Sweden. Other waste categories include working clothes, tools and different kinds of refuse from radiologically controlled areas. Similar types of waste originating from CLAB and Studsvik will also be delivered to SFR-1.

Spent ion exchange resins will be the predominant source of C-14 since the content of C-14 in other waste categories stored in SFR-1 is insignificant. Recently, however, Martin (1986) reported that substantial amounts of C-14 were identified in crud filters from a PWR. According to this single investigation, the amount of C-14 associated with crud was twice the amount retained in the ion exchange resin for the specific power plant examined (PALISADES). PWR plants in Sweden are not equipped with crud filters, and crud is largely retained by the ion exchange systems.

The radiological safety of facilities handling radioactive materials should by law be analysed, and reported to the authorities. A safety analysis report has already been issued and the SFR-1 facility has been approved by the authorities. Since C-14 was one of the nuclides which dominated in the assessment, and since it was also one of the nuclides thought to be evaluated in the most pessimistic way, a special investigation of this nuclide was thought necessary. The radiological consequences of a carbon-14 release are controlled by several parameters.

Essential parameters are:

- source strength
- transport pathways and retention
- biological metabolism

Only the source strength of carbon-14 will be discussed in this context.

Carbon-14 is a radioactive nuclide difficult to measure and great efforts have been made in the past to establish useful correlation factors to other radionuclides measured. One approach has been to use Co-60 as a reference or surrogate nuclide for C-14. EPRI (1980) has recommended the following scaling factors (C-14/Co-60 ratio) in 10CFR61 (=Code of Federal Regulations, Title 10, part 61):

$6 \cdot 10^{-4}$ for BWR and 10^{-2} for PWR (Miller et al., 1985).

Recently, scaling factors of $3.5 \cdot 10^{-2}$ for BWRs and $1.4 \cdot 10^{-1}$ for PWRs have been reported (Cline et al, 1985). In the safety analyses report for SFR-1 a scaling factor of 10^{-3} was applied for both PWR and BWR (SKB 1987).

Although an empirical scaling factor for C-14 with Co-60 might be a most useful tool, numerous detailed studies on the behaviour of C-14 have shown a lack of similarity with any other radionuclide. The concentrations of C-14 in reactor process and waste streams vary greatly from one reactor to another. Nevertheless, major efforts have been made to specify the content of C-14 quantitatively in various kinds of low level waste (Martin, 1986; EPA, 1977; EPRI, 1980; Cline et al, 1985).

The objective in this part of the report is to compare different ways of estimating the C-14 content in spent ion exchange resins.

2 Generation of C-14

C-14 is generated by neutron activation of oxygen, nitrogen and carbon according to:

1: O-17 (n,α) C-14, σ_{th} = 0.24 barns

2: N-14 (n,p) C-14, σ_{th} = 1.8 barns

3: C-13 (n,γ) C-14 σ_{th} = 0.0009 barns

The neutron activation of carbon, reaction 3, plays a less important role. The cross section of carbon is rather small and the source of carbon is mainly present as a minor component in stainless steel. Besides, the isotopic abundance of C-13 in carbon is only 1.1 %.

The other two reactions are far more important. Oxygen is an atomic component in both water and uranium oxide. Nitrogen is an impurity and the concentration level might vary between fuel and reactor types.

C-14 present in fuel elements is not accessible for release during operation.

2.1 Fuel elements

The fuel elements used in Swedish power plants are manufactured by ABB ATOM AB. According to ABB ATOM AB the impurity levels of nitrogen in the cladding material and uranium dioxide pellets are ~ 40 ppm and 5 - 7 ppm, respectively.

The cladding contributes about 10 % of the total weight of a fuel element and the average impurity level of nitrogen in the fuel elements approximates 10 ppm.

The production rate of C-14 in the fuel (exclusive cladding) used in Forsmark 1 and Barsebäck 1 has been calculated elsewhere (Apelquist, 1975; Olsson, 1976). An impurity level of nitrogen in uranium oxide equal to 10 ppm was assumed in the calculation.

Previous results have been adjusted with respect to recent values of nitrogen impurity levels. Typical production rates of C-14 in the fuel elements of Swedish BWRs are given below in Table 2.1.

Table 2.1

Production rate of C-14 in reactor fuel elements of Swedish BWR. Reference plant, Barsebäck 1.

		GBq C-14/GW(e)·year
Uranium dioxide	O-17	189
	N-14	225
Zircaloy cladding	N-14	148
Total		562

Similar calculations have been performed elsewhere. Typical production rates of C-14 in German BWRs and PWRs are 788 and 714 GBq C-14/GW(e)·year (Bonka and Brüssermann, 1974). These results also show that the production rate of C-14 in reactor fuel is of the same order for BWR and PWR.

2.2 Reactor coolant

The production rate of C-14 in the reactor coolant of Forsmark 1 and Barsebäck 1 has been calculated in other contexts (Apelquist, 1975; Olsson, 1976). A small discrepancy was obtained between Forsmark and Barsebäck which could be ascribed to different reactor designs. For the purpose of calculation, the vessel was divided into three nodes, the core volume, the downcomer and the reflector volume. The production rate of C-14 calculated for these different nodes is given below, in Table 2.2.

Table 2.2

Typical production rates of C-14 in reactor coolants of Swedish BWR (Olsson, 1976).

Node	GBq C-14/GW(e)·year
Core	114
Downcomer	100
Reflector	81.4
Total	295.4

The results in Table 2.2 agree very well with corresponding results obtained in other countries. Kunz (1985) reports the average calculated production rates of C-14 in the coolant water of BWRs and PWRs to be 295 and 185 GBq C-14/GW(e)·year, respectively, assuming a concentration of 5 ppm or less of nitrogen in the coolant. The quantity of C-14 generated from 5 ppm nitrogen in the coolant is 10 % of that produced from the oxygen in water. The impurity level of nitrogen in Swedish BWRs is less than 5 ppm and the contribution of C-14 generated from nitrogen has been omitted in the previous calculations presented in Table 2.2 (Olsson, 1976).

2.3 Structures

Carbon is contained in stainless steel and only structures of stainless steel are considered under this heading. Stainless steel corrodes in the water environment of a reactor vessel and the stainless steel surfaces are covered by a protective layer of mainly iron oxides. By different mechanisms, oxides are released to the coolant. The stainless steel quality used is SIS 2333. This quality contains about 0.05 % carbon by weight. The oxide thickness at equilibrium is of the order 100 - 200 mg/dm^2 and the rate of growth is estimated to 2 - 3 mg/dm^2 month. The oxide layer in PWR is mainly composed of magnetite, and in BWR of hematite.

The content of carbon in the oxide layer has been estimated to 0.036 % by weight while the content of oxygen is about 27 % by weight. The production of C-14 from the neutron activation of C-13 and O-17 has been estimated to $2\cdot10^{-1}$ Bq/dm$^2\cdot$year and 30 kBq/dm$^2\cdot$year respectively, assuming an oxide layer thickness of 100 mg/dm^2 and a neutron flux of 10^{12} neutrons/cm$^2\cdot$s. These results show that the contribution of C-14 from the neutron activation of carbon is negligible compared with that from oxygen.

In the reducing environment of PWR it is assumed that C-14 is present as carbides in the oxide layer. Assuming that 25 mg/dm^2 of oxide is released to the coolant yearly and that the activated surface of stainless steel is 100 m^2, about 100-300 MBq C-14 is annually released to coolant. This release of C-14 should be associated with crud. This theoretical value is far below the experimental value reported by Martin (1986). He reports an annual release of C-14 of 120 GBq associated with crud.

3 Transport Pathways

It is generally accepted that C-14 produced in the coolant is released during reactor operation while C-14 formed in the fuel and structural materials is retained. In Sweden the fuel elements containing C-14 will be transferred to CLAB and thereafter to SFL for final disposal.

3.1 Boiling water reactors

A flow scheme of the transport pathways in a BWR is shown in Fig 3.1. In the reactor vessel a fraction of C-14 is carried by the coolant to the primary coolant purification system where C-14 is partly retained by the ion-exchange system. The major fraction of C-14 generated in the reactor vessel is, however, carried by the steam flowing to the turbines. If C-14 is shared between the steam and purification flows according to the flow ratio, approximately 98 percent of C-14 produced in the coolant should be carried by the steam. Carbon species carried by the steam are gaseous and consequently they will be evacuated from the condenser into the off-gas system. Only a minor fraction follows the condensate to the full flow condensate purification system in which C-14 is retained by the ion exchange system. Following purification, the condensate is returned to the reactor vessel as feedwater. The fraction of C-14 retained in the reactor coolant is removed by ion exchange resins in the reactor coolant purification system. The purification system consists of a mixed resin bed. The anion exchange resins are saturated with hydroxyl ions and the cation exchange resins with hydrogen ions.

Numerous measurements of the stack release of C-14 from BWR have been performed abroad and in Sweden. In Germany, it is required by law to measure the gas release of C-14 on a regular basis. In general, the stack releases of C-14 from BWRs vary somewhat irregularly in time and in order to get a true evaluation of the stack release of C-14 it is recommended to make measurements over an extended time period.

The C-14 release from some Swedish power reactors was measured occasionally (Hesböl and Gebert, 1981). The average stack release for five measurements was 420 GBq/GW(e)·year which is somewhat higher compared with the calculated values given in Table 2.2.

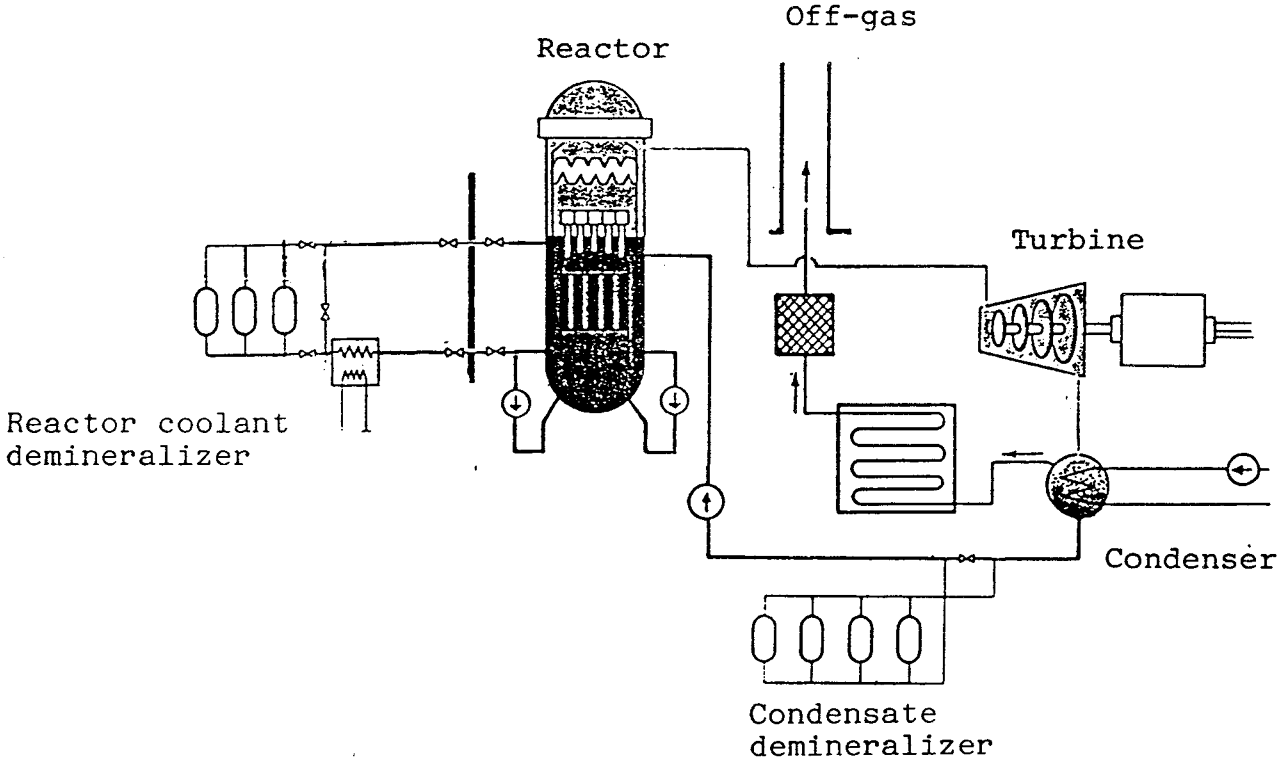

Figure 3.1

Transport pathways and purification systems in BWR.

It has been demonstrated, particularly in Germany and the US, that the stack release of C-14 from BWR corresponds to the production rate of C-14 in the coolant. Based mainly upon his own experiences, Kunz (1985) assumes, in general, that the C-14 releases with liquid and solid wastes are less than 5 % of the gaseous release. Salonen and Snellman (1987) report that less than 1.5 % of the annual gaseous release was contained in the resin waste in the Loviisa PWR. The corresponding figure for the TVO BWR is 0.6 % (Salonen and Snellman, pers comm). With reference to his own investigation, Martin (1986) has estimated a quantity of C-14 in LLW of about 18.5 GBq/GW(e)·year for a nominal BWR, which corresponds to about 5 % of the gaseous release.

At the second IAEA research coordination meetings on C-14 from nuclear facilities in late 1984, Soman et al. (1984) reported that only 60 % of the C-14 produced in the reactor water of Tarapur atomic power station (BWR) was discharged into the air. The residue remained dissolved in the reactor water and a considerable fraction was retained by the ion exchangers of the reactor coolant purification system. Although the fraction of C-14 retained in the reactor coolant was rather high the total gas release was estimated to be in the range 230 - 540 GBq/GW(e)·year for the TARAPUR power plant.

3.2 Pressurized water reactors

A flow scheme of the transport pathways in a PWR is shown in Figure 3.2. The reactor coolant in PWRs is circulated from the vessel to the steam generators and back in three loops. One loop is equipped with a pressurizer to keep the pressure under control. If required, the pressurizer could be vented to the containment. A purification and chemistry control circuit is connected to one of the loops. The flow through the circuit is of the order 10 kg/s (7 kg/s for Ringhals 2) and the circuit is composed of the following subsystems in the order mentioned; a purification system with mixed ion-exchange resins, a fibre filter for particulates, a dumping tank, and a chemical volume control tank. The flow could be directed either to the dumping tank or the chemical volume control tank. The chemistry is controlled by adding feed water and/or boric acid into the chemical volume control tank. The dumping tank and the chemical volume control tank are vented to the gas storage and decay tanks. The coolant is alkaline and the anion exchanger is saturated with borate ions and the cation exchanger with lithium ions.

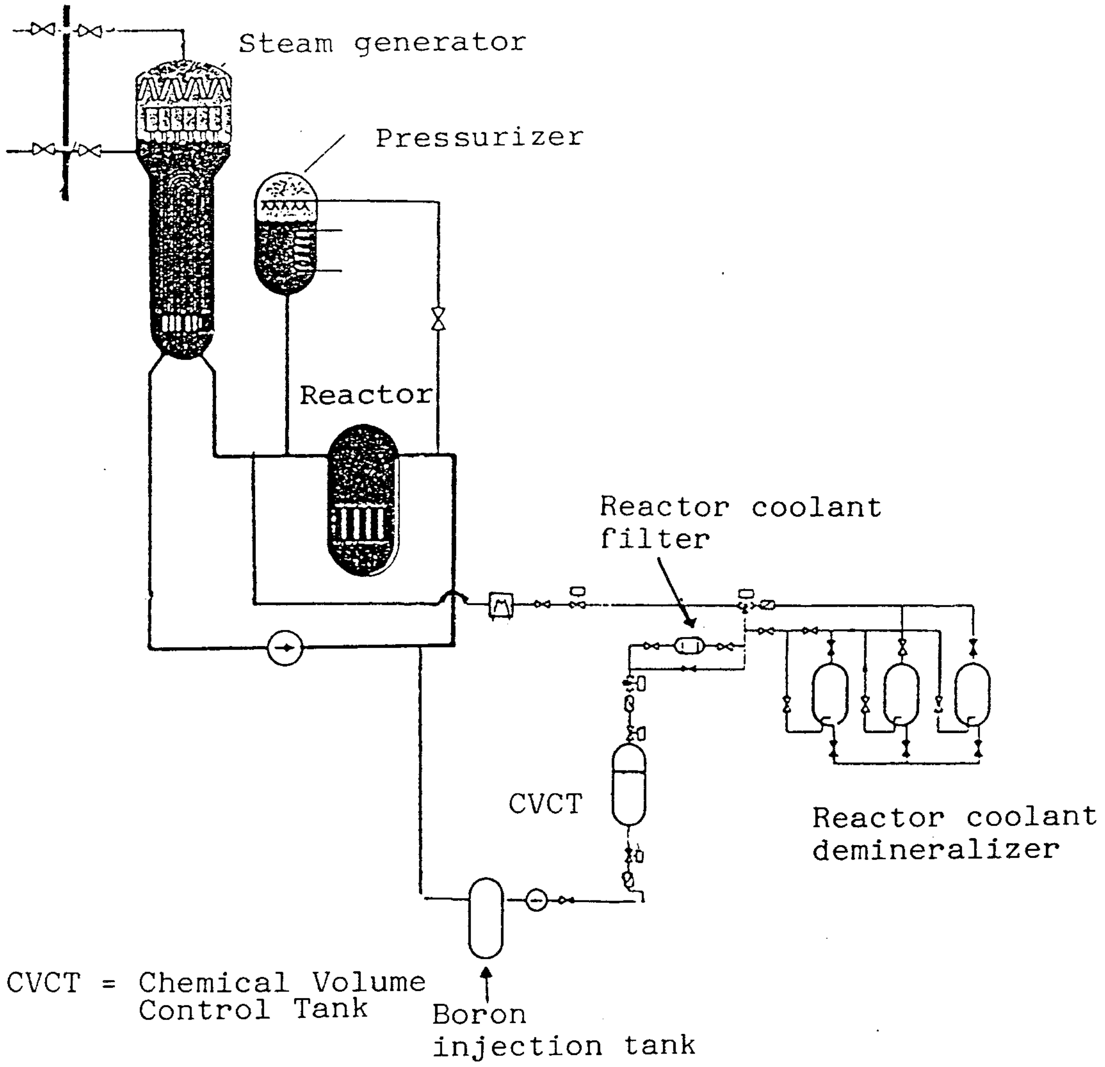

Figure 3.2

Transport pathways and purification systems in PWR.

C-14 is mainly present as organic compounds in the coolant. Only a minor fraction is present as carbon dioxide and carbonates. C-14 is accumulated into the coolant until the removal rate equals the production rate of carbon. The purification flow is rather low and consequently the concentration level of C-14 in the coolant is fairly high ~4000 Bq/kg.

Organic carbon is not retained in the ion exchange system but is vented from the gas space of the dumping tank and chemical volume control tank into the decay tanks. Some volatile C-14 is also vented from the pressurizer into the containment. The minor fraction of carbonate is efficiently retained by the ion-exchange system. The discharge rate of gaseous C-14 from PWR is discontinuous compared to BWR because of the storage and subsequent release of gases from the decay tanks. Even during shutdown, gas discharges might occur.

4 Chemical forms

CO_2, CH_4, C_2H_6, C_3H_8 and C_4H_{10} containing C-14 have been identified in grab samples of stack gas, containment air, off-gas and gas in decay tanks (Kunz, 1985). It is generally accepted that these chemical forms of carbon and even carbon monoxide are included although the relative abundance of different chemical species might vary from one reactor to another. Hence sampling equipment for gas-sampling of C-14 in general is designed to trap carbon dioxide and organic carbon. In principle, carbon dioxide is absorbed in an absorber, liquid or solid. Organic carbon and carbon monoxide penetrating the absorbers are oxidized catalytically in a furnace to carbon dioxide which in turn is absorbed in a subsequent absorber. In this way the chemical forms of C-14 are classified as carbon dioxide and organic carbon. Among the organic forms, methane dominates according to gas chromatographic analysis (Kunz, 1985). In BWRs applying normal water chemistry the gas discharges of carbon dioxide are overwhelming. The fraction of organic carbon is of the order 0.05.

For some time trials have been performed to operate BWRs at HWC conditions (HWC = hydrogen water chemistry.) An introduction of hydrogen leads to reducing conditions and the generation of organic carbon is favoured. PWR is operated at reducing conditions and correspondingly the generation of organic carbon is favoured compared with carbon dioxide. The fractions of organic carbon obtained for several power reactor units in a Swedish investigation are shown in Table 4.1 (Hesböl and Gebert, 1981).

Table 4.1

Fractional release of organic carbon from some Swedish power reactor units.

Reactor unit	Fractional release of organic carbon
Oskarshamn 1 BWR	0.03
Oskarshamn 2 BWR	0.06
Barsebäck 2 BWR	0.05
Ringhals 1 BWR	0.19 (1978)
Ringhals 1 BWR	0.46 (1980)
Ringhals 2 PWR	0.98 (1978)
Ringhals 2 PWR	0.58 (1979)

5 Retention of C-14 in ion exchange resins

The chemical forms of carbon in the coolant are most likely carbon dioxide and organic carbon. The ratio between these types of chemical forms depends on whether the chemical conditions are oxidizing or reducing. The organic carbons (alkanes) are inert forms and will not be trapped by ion-exchange resins. Carbon dioxide dissolves in water according to Henry's law. A fraction is hydrolysed and dissociated into bicarbonate or carbonate ions which are retained in anion exchange resins. In fact it has been demonstrated that C-14 in reactor coolants is trapped by anion exchange resins and not by cation exchange resins. Isotope exchange reactions with the organic resin itself seem to be negligible (Salonen and Snellman, 1987).

Bicarbonate ions are predominant in the BWR reactor coolants. Bicarbonate anions are weakly bound to the exchange resins. The selectivity coefficient of bicarbonate relative to chloride on Dowex 1 is 0.32 while the selectivity coefficient of hydroxide is 0.09. Bicarbonate ions are thus readily exchangeable with hydroxide ions. The affinity of bicarbonate ions to the resin is fairly good. The residence time of bicarbonate ions is roughly one-third of the residence time for chloride ions.

In the following, the retention of C-14 in ion exchange beds is discussed on an experimental basis. The experimental works referred to have been published quite recently (Martin, 1986; Kunz, 1985; Soman et al., 1984; Salonen and Snellman, 1987).

5.1 Pressurized water reactors

Kunz (1985) has examined the removal of C-14 by ion exchange resins by taking liquid grab samples before and after the clean-up demineralizers. The results of the analysis showed no detectable removal of C-14 by the demineralizer. Two power units were investigated.

The chemical forms of the gas discharges from PWRs are mainly organic carbons. Organic carbons are also expected to be predominant in the primary coolant. In fact, Salonen and Snellman (1987) have shown that only 0.3 % of C-14 was in the form of CO_2 in the reactor water of Loviisa 1. Organic carbons (alkanes) remain undissociated in water, hence, organic carbons are not retained by the ion exchange resins. This is probably the reason why Kunz (1985) could not detect any removal of C-14 by the demineralizers. The fraction of C-14 in the form of CO_2 is, however, retained by the ion exchangers. Salonen and Snellman (1987) have shown that in a mixed bed C-14 is retained by the anion exchange resin and hardly at all by the cation exchange resin.

The content of C-14 in spent resins from PWR can be calculated if the concentration of C-14 as carbon dioxide in the coolant, the coolant purification flow rate, the operational time of the demineralizers and the removal efficiency of the demineralizers are known. Another approach is to estimate the loading of C-14 in each kg of ion exchange resin and take this number times the annual consumption of ion exchange resins. Both approaches have been reported earlier (Kunz, 1985; Salonen and Snellman, 1987). Loss of C-14 during post treatment of ion exchange resins should be considered. Salonen and Snellman (1987) have shown that the loss of C-14 during bituminization could be as high as 97 %.

5.2 Boiling water reactors

In BWR the only ionic species of C-14 is bicarbonate. Even carbon dioxide dissolved in the coolant can be considered dissociated into bicarbonate ions when passed through an ion exchange column. Kunz (1985) has measured the decontamination factor for reactor coolant demineralizers by taking grab samples of the liquid before and after the demineralizers. On one occasion he obtained a decontamination factor of 7, indicating that 87 % of C-14 was retained by the ion exchange clean-up system. A second attempt to measure the decontamination factor failed because of the very low concentrations of C-14 in the coolant before the demineralizer. The detection limit of C-14 in the coolant was 0.5 Bq/kg and the concentration of C-14 in the reactor coolant was less than the detection limit. The concentration of C-14 in the reactor coolant on the first occasion was about 70 Bq/kg and the removal of C-14 by the reactor coolant clean-up demineralizer varied by more than two orders of magnitude. Salonen and Snellman (1987) report low concentrations of C-14 in the reactor coolant, 1 - 2 Bq/kg, while Soman et al. (1984) reports concentration levels of ~170 Bq/kg.

In BWR C-14 is expelled almost completely from the condenser into the off-gas stream. However, some C-14 by-passes the condenser into the condensate. In fact, Salonen and Snellman (1987) report concentration levels of 2 - 4 Bq/kg condensate which is somewhat higher than the concentration in the reactor coolant. They also report loadings of 73 kBq/kg dry weight for reactor coolant clean-up demineralizer and 1.5 - 120 kBq/kg dry weight for condensate demineralizer. Martin (1986) reports a loading of 140 kBq/kg for the reactor coolant clean-up demineralizer. Neither he nor Kunz expected any substantial removal of C-14 by the condensate demineralizer. Kunz reports that no C-14 decontamination could be detected to take place in the condensate demineralizer.

5.3 Retention of crud in ion exchange resins

Ion exchange resin beds of the type used for reactor coolant and condensate clean-up are excellent crud filters. In fact, ion exchange resins have to be replaced because of unacceptable pressure drops across the filter rather than poor ion exchange capacity. C-14 associated with crud has not been reported for BWRs, only for PWRs. The reactor coolant purification system in Swedish PWRs is equipped with a particle filter downstream of the ion exchange resin bed. This particle filter should be considered as a backup filter for ion exchange resin particles rather than a crud filter. Thus in Swedish PWRs C-14 associated with crud particles is expected to be retained in the ion exchange resin beds.

6 Source strength estimations

The only source of C-14 in the waste from nuclear power plants is spent ion exchange resins. C-14 is chemically bound as bicarbonate or carbonate ions to the resins. In resins from PWR some C-14 can be present in crud particles, probably as carbides. Three different approaches have been applied in the following to estimate the source strength of C-14 in ion exchange resins. The first approach applies a scaling factor, the second a decontamination factor and the third a loading factor to estimate the source strengths.

6.1 Scaling factors

C-14 is a difficultly measured nuclide and a convenient method to estimate the C-14 content is to relate the activity of C-14 to the activity of an easily measured nuclide. Co-60 seems to be a suitable reference and numerous analyses have been performed in different laboratories to specify a reliable scaling factor. The scaling factor is defined as the ratio between the activities of C-14 and Co-60. Scaling factors ranging from 10^{-2} down to $6 \cdot 10^{-4}$ have been reported from different laboratories. Scaling factors are convenient tools, but should be related to the type of waste. It is believed that different scaling factors should be applied for resins from reactor coolant clean-up systems of PWR or BWR and condensate clean-up systems in BWR. If the true scaling factor depends on the type and relative amounts of waste, great care should be taken in selecting a relevant scaling factor. A scaling factor of 10^{-3} has been applied in the SFR-1 safety analysis report. This value is in between the extremes of reported literature values. The results reported here have been taken from the SFR-1 safety analysis report (SKB, 1987).

6.2 Decontamination factors

The retention of C-14 in an ion exchange bed can be calculated from the concentration of C-14 in the liquid before the ion exchange column and the decontamination factor.

6.2 1 PWR

The concentration of C-14 in the reactor coolant before entering the ion exchange bed in PWRs is of the order 4000 Bq/kg. There is good agreement between results obtained in Finland (Salonen and Snellman, 1987) and the US (Kunz, 1985). The decontamination factor is very low because of the high fraction of organic carbon. Considering only carbonate ions, the decontamination factor is fairly high. The fraction of CO_2 in the reactor coolant of Loviisa 1 was 0.3 %. In general, a reasonable fraction of carbonate ions in PWR coolant seems to be of the order 1 %. The max. retention of C-14 in the ion exchange resins of PWR has been calculated to be 9 GBq C-14/GW(e)·year assuming a purification flow of 7 kg/s (Ringhals 2) and complete removal of C-14 in the ion exchange system. The concentration of exchangeable C-14 in the coolant was assumed to be 40 Bq/kg. According to Martin (1986), 122 GBq C-14/GW(e)·year should be added as a contribution of C-14 associated with crud. C-14 in crud has so far not been confirmed by any other investigators.

6.2.2 BWR

Different concentration values of C-14 in the reactor coolant have been reported. Kunz (1985) reports two values which differ in magnitude with an order of two. Only two measurements were made; the lowest being 0.5 Bq/kg. Corresponding values reported by Salonen and Snellman (1987) for TVO 1 were also fairly low. The concentration level was of the order 1 - 3 Bq/kg. Soman et al. (1984) report that a considerable fraction of C-14 remained dissolved in the reactor water. Analysis of liquid grab samples taken before and after the ion exchange system gave average concentrations of 166 Bq/kg before and 12 Bq/kg after the clean-up system. Accordingly, the removal efficiency of the ion exchange system is better than 90 %. The decontamination factor was 14. The power reactor unit investigated was Tarapur Atomic Power Station, a BWR with an electrical output of 150 MW.

For the time being a best estimate of the concentration level of C-14 in the reactor coolant is 2 Bq/kg.

The retention of C-14 in the reactor coolant demineralizers has been estimated to be 1.6 GBq/GW(e)yr assuming a purification flow of 25 kg/s (Forsmark 1 and 2) and complete removal of C-14 in ion exchange resins. Kunz (1985) reports a decontamination factor of ~7 for the reactor coolant resin system. Very few data on the removal of C-14 in the condensate demineralizers exist. Kunz reported that the removal of C-14 in the condensate demineralizers of Fitzpatrick was not detectable. In fact only Salonen and Snellman (1987) report data on concentration levels of the condensate and contents of C-14 in the condensate demineralizers. They report a concentration level of 2 - 4 Bq/kg condensate. A concentration level of 3 Bq/kg has been applied in the calculations.

The retention of C-14 in the condensate demineralizers has been estimated to be 130 GBq/GW(e)yr assuming a condensate flow of 1350 kg/s (Forsmark 1) and complete removal of C-14 in the demineralizer. The retention of C-14 in the condensate demineralizers is one-third of the stack release. The condensate concentration level or the decontamination factor applied thus seems to be too high.

6.3 Loading factor

The loading factor is defined as the activity of C-14 per kg of ion exchange resin dry or wet as specified. Loading factors have been reported by several authors. Some of them are listed below.

Table 6.1 Loading factors for C-14 on ion exchange resins.

Reference	PWR (kBq/kg)	BWR (kBq/kg)
EPRI (1980)	18.5 a)	1.1 b)
EPA (1977)	185	7.4 b)
Martin (1986)	1924	140 b)
Salonen and	690-1770	73 b)
Snellman (1987)		1.5-120 c)

a) Fuel pit resin
b) Reactor coolant demineralizers
c) Condensate demineralizers

The spread of data is large. The most recent data, those of Martin and Salonen (1987), do not deviate too widely for the reactor coolant demineralizers. The Finnish investigators have only made two measurements of the condensate demineralizers and the data reported in

Table 6-1 are the results of these measurements. In his calculations, Martin (1986) uses a density equal to 1 g/cm^3 for the resin, which would correspond with the density of wet resin, while Salonen refers their values to dry resin (density ~ 0.5g/cm^3). Table 6.2 presents the best estimates of the parameters used in the calculations.

Table 6.2

Loading factors and annual consumption of ion exchange resin applied in the calculations.

	Loading factor kBq/kg	Annual amount of resin kg/GW(e)·year
PWR		
coolant	2 000	1 000
BWR		
coolant	140	3 000
condensate	120	13 000

The source strengths of C-14 on a yearly basis in ion exchange resins from different systems based upon the information given in Table 6.2 are presented in Table 6.3.

Table 6.3

The source strenghs of C-14 in spent ion exchange resins according to loading factors.

	GBq/GW(e)·year
PWR	2
BWR	
reactor coolant demineralizers	0.4
condensate demineralizers	1.6

7 Conclusions

Three different approaches have been reviewed to obtain a best estimate of the content of C-14 in spent ion exchange resins from commercial nuclear power plants. The first approach applies a scaling factor, the second a decontamination factor and the third a loading factor. A compilation of the results is given in Table 7.1.

Table 7.1

The content of C-14 in spent resins calculated in three different ways.

	GBq C-14/GW(e)·year Scaling factor	Decont. factor	Loading factor
PWR	24	9	2
BWR	24	132	2

The values under the heading "scaling factor" are derived from the source strength of C-14 at year 2010 reported in the safety report of SFR-1 (SKB, 1987). The same scaling factor, 10^{-3} was used for both PWR and BWR waste in the safety report of SFR-1. The value 132 under the heading "decontamination factor" in Table 7.1 does not fit in with the others. The C-14 in the condensate demineralizers contributes with 130 GBq/GW(e)·year which seems to be far too high. The value corresponds neither with the values derived from the scaling factor nor with the loading factor.

On the whole, there are very few experimental data and the spread of existing data is very large. Obviously there is a great need for more experimental data to improve the statistics. The chemistries of Co^{++} and C-14 are so different, and the application of the same scaling factor for all applications does not seem to be recommendable. To treat each purification system separately, either by applying specific scaling factors or other types of analysis, seems more reliable.

It has recently been feared that C-14 activity associated with crud could be an important source of C-14 in PWR. Calculations have shown that C-14 in crud could contribute about

100 MBq/GW(e)·year, while estimations based upon experimental observations are of the order 100 GBq/GW(e)·year. Confirmatory measurements are obviously needed.

Isolation of Carbon-14 in underground repositories in granitic bedrock

1 Introduction

The relative importance of ^{14}C as a radiation source from both nuclear explosions and nuclear power production is documented by UNSCEAR (1982). In nature ^{14}C is produced in the upper atmosphere by the reaction of ^{14}N with secondary neutrons from cosmic rays. The production rate from this natural source is approximately 10^{15} Bq/y, giving a global inventory of 8500 PBq ($8.5 \cdot 10^{18}$ Bq) and a specific activity both in the atmosphere and in living organisms of 227 Bq/kg of carbon (UNSCEAR, 1982; Killough, 1980; Bush et al., 1984). The natural production of ^{14}C in U and Th ores is discussed by Jull et al. (1987).

From the half life of ^{14}C (5730 y) the following relationships are found

$$2.31 \cdot 10^{12}\ \text{Bq-}^{14}\text{C} = 1\ \text{mol-}^{14}\text{C}$$

$$1.65 \cdot 10^{11}\ \text{Bq-}^{14}\text{C} = 1\ \text{g-}^{14}\text{C}$$

$$3.73 \cdot 10^{10}\ \text{Bq-}^{14}\text{C} = 1\ \text{g-}^{14}\text{CO}_3^{-2}$$

$$3.67 \cdot 10^{10}\ \text{Bq-}^{14}\text{C} = 1\ \text{g-H}^{14}\text{CO}_3^{-}$$

For example, a water containing 0.3 mg/l (ppm) of ^{14}C-bicarbonate, has an activity of 0.011 GBq/l.

Studies on the consequences of possible leakage from the Swedish repository for Low and Intermediate Level Nuclear Reactor Waste (SFR) have identified ^{14}C as one of the important nuclides in the inventory (Bergström and Puigdomenech, 1987; Wiborgh et al., 1987b) in agreement with other studies (Pigford, 1984; Malbrain and Lester, 1987; Oversby, 1987; Burkholder et al., 1976; Vieno et al., 1987; Häggblom and Kjellbert, 1979).

Carbon-14 is of special concern because of its long half life (5730 y) and because it is taken up by plants from the atmospheric carbon dioxide during photosynthesis, being thus incorporated in the human food chain. Human tissues reach equilibrium with atmospheric ^{14}C with a delay of 1.4 y (UNSCEAR, 1982).

This part of the report contains a literature survey and a discussion of the processes occurring in a geologic repository. Similar reports are those of Bush et al. (1984) and Liepins and Thomas (1988).

The part of the report is written with special emphasis on the concrete silo in the SFR repository, which is situated at Forsmark (on the Baltic Sea coast, about 110 km north of Stockholm) approximately 50 meters below the rock surface.

In order to be able to present quantitative results, in this report we will use the values in Table 1.1 as a reference model repository. The values are selected to simulate the silo in the SFR reactor waste repository at Forsmark, and therefore, most of the data in Table 1.1 have been adapted from Wiborg and Lindgren (1987a) and Neretnieks et al. (1987). As discussed by Bergström and Puigdomenech (1987), the highest calculated radiological consequences from any radionuclide leakage, correspond to a scenario where the underground repository remains intact for 2500 years. During that time the rising of the Scandinavia peninsula will lift the repository site above the Baltic sea surface, and the radionuclides might be discharged directly either to a well, a small lake, or to agricultural land.

The initial amount of carbon-14 in the repository is assumed to be 5 TBq, which is close to the amount expected for the Swedish production by the year 2010 (this report, Part 1), after it has decayed for 2500 years.

Table 1.1

Description of the model repository discussed in this report that simulates the silo in SFR at Forsmark.

^{14}C inventory (at 2500 y after repository closure):		5 TBq
Amounts of Materials		
Concrete:	- silo structrure	4632 m^3
	- internal walls	6971 "
	- porous concrete (as backfill in 67 compartments)	3350 "
	- walls of concrete boxes	> 4413 m^3
	- conditioning concrete	~ 7170 "
Total:		~ 26530 m^3 (= $8.62 \cdot 10^6$ kg)
Plastics (ion exchange resins):		$(8.4 - 6.9) \cdot 10^6$ kg wet resins = $(3.3 - 2.8) \cdot 10^6$ kg dry resins
Bitumen:		$(1.4 - 1.2) \cdot 10^6$ kg
Material Properties		
Concrete composition:		325 kg cement/m^3
		64.5 % CaO (= 3740 mols/m^3)
		0.8 % K_2O (= 27.6 ")
		0.2 % Na_2O (= 10.5 ")
Porosities:	host rock	10^{-4} m^3/m^3 (= 0.1 %)
	concrete	0.20 "
	clay/sand	0.61 "
	void space above silo (filled with crushed rock)	0.30 "
	waste containers	0.20 "
Repository size		
Rock cavity:		
	Height	5.0
	Width	32.4
	Depth	23.6
	Total Volume	49702 m^3

Table 1.1 (Continued)

Concrete structure (silo):			
	Height	50.0 m	
	Width	28.4	
	Depth	23.6	
	Wall thickn.	0.8	
	Total volume:	35400 m^3	
Clay/Sand backfill:			
	Thickn. top & bottom	2.5 m	
	" sides	1.2	
	Total volume:	3328 m^3	
Void space above concrete structure (filled with crushed rock):			
	Height	10.0 m	
	Width	32.4	
	Depth	23.6	
	Total volume:	7646 m^3	
Hydrology			
Water flux in	host rock	5 l/(m^2 y)	horizontal
" "	rock around silo	10 "	"
" "	inside silo	10 "	vertical upward
" "	in clay/sand walls	30 "	" "
" "	void space above silo (filled with crushed rock)	15 "	horizontal
		0.5 "	vertical
" "	fracture in the granite	70 "	horizontal

2 Chemical forms of carbon-14 resulting from nuclear power production

Carbon-14 is produced in nuclear power plants mainly by neutron activation reactions of ^{17}O (present in the oxide fuel and in the moderator) and ^{14}N present in the fuel as an impurity (Bush, et al., 1984; Bush, 1984). Normal effluent release of ^{14}C from reactor operation is mainly as $CO_2(g)$ (> 95 %) for BWR reactors and CO, $CH_4(g)$ and other hydrocarbons for PWR reactors (UNSCEAR, 1982; Bush et al., 1984; Kahn et al., 1979). Methane released to the atmosphere has a turnover time of about 5 y and is distroyed by radical reactions in the stratosphere (Enhalt, 1974; Pearman and Fraser, 1988).

In the spent fuel elements carbon will be present as carbides, elemental carbon or as entrapped carbon oxides or methane. The amount of ^{14}C in spent fuel is uncertain (Van Konynenburg et al., 1987; Bechthold et al., 1987) and is discussed elsewhere (this report, Part 1).

The chemical stability of any carbides present in spent fuel towards hydrolysis depends on the other element involved (Greenwood and Earnshaw, 1985). For example, interstitial carbides of La, U, Th, Pu, Ti, Zr, Hf, V, W, etc, do not react with water, while salt carbides (e.g., of Be, Al, Ca, Na, etc) hydrolyze to form methane, or ethyne. Other types of carbides (e.g., Fe, Co, Ni, lanthanoids and actinides) react with water to form diverse hydrocarbons.

In reprocessing plants, more than 75 % of the ^{14}C inventory is released from the fuel in the form of $CO_2(g)$ (UNSCEAR 1982). Carbon-14 is also released as CO_2 from fuel elements in air or in aqueous solutions, but the release of ^{14}C is small in the absence of oxygen, indicating that carbon-14 might be present as elemental carbon in the spent fuel (Angus et al., 1986; Van Konynenburg et al., 1987 and 1985; Wilson and Shaw, 1987). It must be noted that both CO and CH_4 would react with O_2 to form CO_2 in the radiation field of spent fuel (Van Konynenburg et al., 1985).

Carbon-14 contained in Low and Intermediate Level Nuclear Waste mainly originates in the nuclear fuel cycle. The highest ^{14}C activities are found on filter cartridges, filter sludges and ion exchange resins (Gruhlke et al., 1986). The chemical form is not well known, but it is usually assumed that it is mainly present as carbonate (Gruhlke et al., 1986). Very few carbon-14 analyses of ion exchange resins have been reported in the literature (Snellman and Salonen, 1982; Salonen and Snellman, 1987; Cline et al., 1985). There is a rather large variation between the different analyses reported. Snellman and Salonen (1982) analyzed Finnish ion exchange resins for both $^{14}CO_2$ (after acidification) and ^{14}CO and hydrocarbons. They found that most of the carbon-14 in CO and hydrocarbons escapes from the resins during normal handling operations, and that 93 to 99 % of the radiocarbon remaining in the resin is bound to CO_2. The amount of ^{14}C that might escape from the resins will depend on the handling procedure and matrix used for the waste.

3 Physico/chemical processes affecting ^{14}C in the repository and in the geosphere

The changes expected to take place in a repository are due to interactions between the different materials and between repository components and groundwater. These processes may be summarized as follows (Biddle and Rees, 1988):

- radiolysis will deteriorate ion exchange resins and contribute to gas formation
- microbial degradation of organic materials will form soluble organic carbon species and will produce some gas
- corrosion of metals will produce the larger gas quantities
- concrete and bentonite degradation will affect the hydraulic properties of the repository
- other chemical reactions will degrade ion exchange resins, precipitate carbonates, etc

In the geosphere, microbial activity will induce changes in the dissolved organic contents of the groundwater, and affect precipitation/dissolution of calcite as a fissure filling mineral. Sorption, diffusion and hydrodynamic dispersion will affect the transport of ^{14}C in the rock matrix.

3.1 Bacterial activity

Several reviews on the effects of microbiological activity on nuclear waste management are found in the literature (Colombo et al., 1982; West et al., 1982 and 1985; Loewen and Flett, 1984; Francis 1985; McGahan, 1987). As carbon is an element present in all living organisms, it is possible (at least in principle) that part of the ^{14}C activity of the repository is taken up by microorganisms. This might affect the mobility of ^{14}C. For spent fuel repositories, the available amounts of nutrients and carbon are not as high as in low level waste repositories, and therefore, our discussion will be centered on LLW and MLW repositories.

The mobility of ^{14}C is affected by bacterial activity, for example, microorganisms might migrate from the repository, carrying away ^{14}C-organic compounds in their bodies. Furthermore, the anaerobic bacterial degradation of the organic compounds present in the repository will also produce a diversity of organic chemicals. Reported dissolved organic carbon from trench leachates and well water from LLW disposal sites has values between 2 and 7 000 mg/l (Francis et al., 1980b; Dayal et al., 1986; Husain et al., 1979). These organic compounds, together with bacterial remains, might contain ^{14}C and will be transported by groundwater as dissolved organic carbon.

Two other possible products of bacterial metabolism that also contain ^{14}C and will easily migrate are: carbon dioxide and methane. For example, it has been found that $^{14}CH_4$ emanates from shallow land burial trenches of LLW (Colombo et al., 1982; Francis, 1984; Husain et al., 1979; Kunz, 1982; Francis et al., 1980a). The bacterial formation of gas is further discussed in section 3.2.

Heterotrophic bacteria can only obtain energy and structural carbon from organic compounds found in their environment. On the other hand, autotrophic bacteria use inorganic carbon to produce their own organic constituents, and take energy from the oxidation of reduced compounds.

As the physico/chemical conditions in most repositories are such that the initial $O_2(g)$ present on repository closure is quickly consumed, only anaerobic microorganisms are of importance in this context. Under anaerobic conditions (absence of oxygen), autotrophs take energy by indirect oxidation through dehydrogenation. Methanogenic bacteria are autotrophic microorganisms that oxidize hydrogen (Colombo et al., 1982):

$$CO_2(aq) + 4H_2 \rightarrow CH_4 + 2H_2O$$

This reaction allows bacteria to generate $^{14}CH_4$ and carbon-14 organic compounds from inorganic ^{14}C in $H^{14}CO_3^-$, $^{14}CO^{2-}$ and $^{14}CO_2(aq)$. Francis (1984) reports the bacterial generation of $^{14}CH_4$ from trench leachates in laboratory experiments.

Other substrates like acetate, formate and methanol are also bacterially reduced to methane, for example,

$$CH_3COO^- + H_2O \rightarrow CH_4 + CO_2(aq)$$

Nevertheless, autotrophic bacteria need nitrogen and phosphorus for their protein formation. The nitrogen source might be ammonium, which is often present in LLW repositories, and therefore, it is generally believed that the requirements for growth of methanogenic bacteria are usually met in LLW repositories (Colombo et al., 1982).

In environments where pH>8 (for example in cement repositories) methane production might not occur because methanogenic bacteria do not tolerate such alkaline environments (Colombo et al., 1982, p 31). However, other types of microorganisms (e.g. dentrifying bacteria) well tolerate alkaline environments.

Many other reactions are also catalyzed by bacteria, for example (Wetzel, 1983, p 237 & 324):

$$SO_4^{2-} + 2(CH_2O) + 2H^+ \rightarrow H_2S + 2CO_2(g) + 2H_2O$$

$$2NO_3^- + 1/6(C_6H_{12}O_6) \rightarrow 2NO_2^- + CO_2(g) + H_2O$$

Other reactions can be found in the literature (Colombo et al., 1982; Stumm and Morgan, 1981, p 458; Loewen and Flett, 1984).

Methylotrophic organisms are bacteria (and some yeasts and fungi) that oxidize CH_4 (Colombo et al., 1982; Wetzel, 1983, p 601). These organisms catalyze the reaction:

$$5CH_4 + 8O_2 \rightarrow 2(CH_2O) + 3CO_2 + 8H_2O \qquad \text{Eq 3.1.1}$$

which will reduce the release of methane that is produced in a repository to the biosphere.

3.2 Microbial Gas Production

Literature references on the bacterial decay of plastic materials and bitumen are very scarce (Biddle et al., 1987 and McGahan, 1987). The only available experimental data for gas release from plastic material appears to be the studies of Bowerman et al. (1988) and Molecke (1981). Barletta et al. (1987) and Wolf (1989) also report on the biodegradation of bitumen. The decay of other organic chemicals found in trench leachates is reported by Francis (1982 and 1984).

The rate of gas production due to bacterial activity under anaerobic conditions in nuclear waste repositories has also been reviewed in Rennerfelt (1981), Biddle et al., (1987) and Lever and Rees (1987). The data given in the reviews by Molecke (1981) and Rennerfelt (1981) are often used to calculate microbial gas production in nuclear waste repositories (e.g., Wiborg et al., 1983; NAGRA 1983; Snellman and Uotila, 1985; Zuidema and Höglund, 1988).

The values given by Rennerfelt (1981) are based on the gas production rates of Molecke (1981) and are summarized in Table 3.2.1. The figures indicate that all ion exchange resins will be degraded in 4 000 years, and that all bitumen will have disappeared in 10 000 years. It is difficult to judge the validity of this assumption, because it is not based on any experimental data. The degradation of the active sites is expected to occur faster than that of the three dimensional polystyrene structure. As mentioned on p 2:9 (section 3.1). it is possible that the high pH environment in concrete repositories may prevent some bacterial growth, and therefore, microbial gas production from ion exchange resins and bitumen is often assumed to be negligible. This approximation was used both by Wiborgh et al. (1983) and Moreno and Neretnieks (1988) in their calculation of gas generation in the silo of SFR at Forsmark.

Table 3.2.1

Bacterial gas production (Rennerfelt, 1981).

Degradation of	mol gas / kg	
Bitumen:	22.2	CO_2
(10 % degrad in 1000 y)	50.1	CH_4
Ion exchange resins	29	CO_2
(5 % degrad in 200 y)	48	CH_4

If we assume, nevertheless, that bacterial gas production is not hindered by high pH values in a cement environment, the following calculations on microbial gas production may be made.

Using data in Table 1.1, the amount of resins is about $3.6 \cdot 10^6$ kg and that of bitumen ~ $1.4 \cdot 10^6$ kg. If we use as an example the rates given in Table 3.2.1, after 4 000 years $277 \cdot 10^6$ mol gas would have been generated by the degradation of the resins ($=6.2 \cdot 10^6$ m^3 NTP). Similarly, after 10 000 y, the total amount of gas generated by the degradation of bitumen would be $101 \cdot 10^6$ mol gas ($=2.3 \cdot 10^6$ m^3 NTP). The potential gas generation by bacteria is therefore $3 \cdot 10^6$ m^3 CO_2 and $5.5 \cdot 10^6$ m^3 CH_4. If all 2.17 moles of ^{14}C initially present in the repository are associated with the ion exchange resins, the initial ^{14}C specific activity

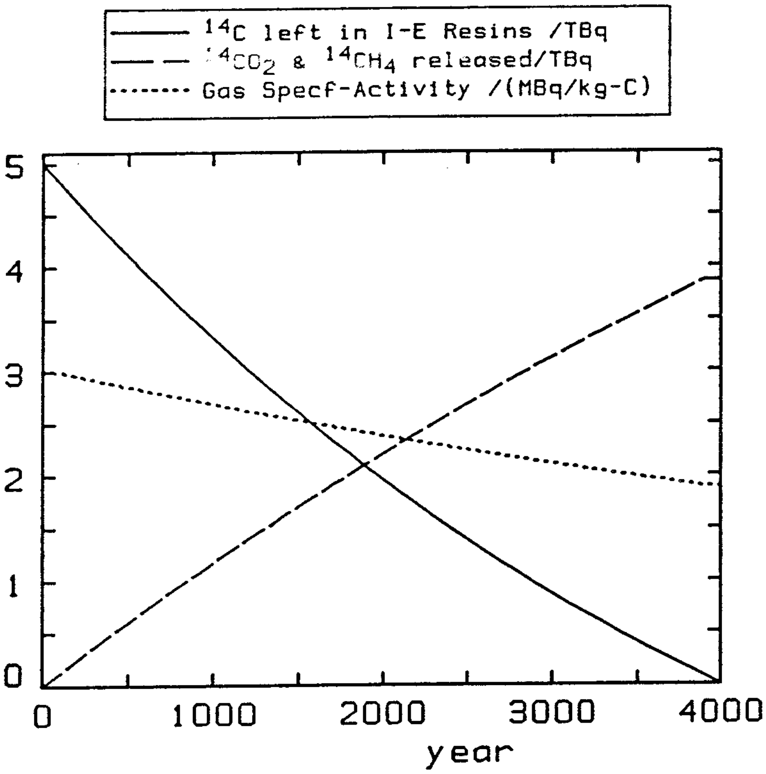

Figure 3.2.1

Carbon-14 release through bacterial gas production. The calculations were made assuming that rates of gas generation given in Table 3.2.1 are correct. The amounts of ion-exchange resins and bitumen given in Table 1.1 were used. The figure shows the amount of ^{14}C left in the ion-exchange resins, and released in the CO_2 and CH_4 produced by microorganisms. The specific activity of the gases is also given (in MBq/kg-carbon).

in the gas generated would be 3 MBq/kg-carbon, the initial amount of carbon-14 released through this mechanism ~1.3 GBq/year, and the total amount of ^{14}C-released from the repository is 3.9 TBq (during the 4000 year period during which the ion exchange resins are assumed to be totally consumed by bacteria). This hypothetical gaseous release of carbon-14 is illustrated in Figure 3.2.1. If the gas generation rate (from metal corrosion, radiolysis and bacterial activity) is about 10^4 m^3/year, the initial ^{14}C-activity of the emanating gas would be $1.3 \cdot 10^5$ Bq/m^3.

Gas transport by advection and diffusion are expected to be small as compared with the rates of gas generation, and it is usually believed that it will escape to the atmosphere through fractures in the rock matrix (Thunvik and Braester, 1986; Ross, 1988). The radiological impact of such a gaseous release will depend on the primary recipient, which in the case considered here (SFR at Forsmark) might be the Baltic sea, a small lake area, agricultural land or a housing area.

The figures given above for gas release of carbon-14 are overestimates because:

- some carbon dioxide and methane will be transported dissolved in groundwater by diffusion and advection
- some $^{14}CO_2$ will react with the calcium hydroxide of concrete (Dayal and Klein, 1988) and will be retained as carbon-14 calcite (cf. eqn 3.2.1)
- part of the methane production will be used by methylotrophic organisms in the soil layer or sediments of the gas discharge zone, as indicated in reaction 3.1.1

It might be argued that the release of ^{14}C from the ion exchange resins might be faster than the bacterial decomposition of the resin's matrix (because decay of the reactive groups is faster than that of the three-dimensional plastic structure), and that therefore the initial ^{14}C concentration in the gases released from the repository might be higher. However, in a cement environment, the reaction

$$Ca(OH)_2(c) + CO_3^{2-} + 2H^+ \rightarrow CaCO_3(c) + 2H_2O \qquad \text{Eq 3.2.1}$$

which will be in equilibrium at repository closure, will be shifted to the right because of the bacterial production of CO_2. If $^{14}CO_3^{2-}$ is released faster from the resins than non-active carbonate species, it will also be selectively retained by calcium carbonate precipitation in the surface of the cement matrix.

3.3 Chemical reactions in concrete

Large amounts of concrete are present in the Swedish SFR repository for LLW and MLW (c.f., Table 1.1).

This is an important factor because carbon-14 carbonate species are known to be retained by concrete. Literature K_D-values are given in Table 3.3.1. Furthermore, carbonate solubility will be low in a concrete environment, due to the reaction

$$\underset{\text{portlandite}}{Ca(OH)_2} + CO_3^{2-} + 2H^+ \rightarrow \underset{\text{calcite}}{CaCO_3} + 2H_2O \qquad \text{Eq 3.2.1)}$$

$$\log K_{eq} = 31.1 = 2pH - \log \{CO_3^{2-}\}$$

This equation might be used to estimate maximum levels of dissolved inorganic carbon, for example, at pH=13, $\{CO_3^{2-}\} = 8 \cdot 10^{-6}$ mol/l, while at pH=11, $\{CO_3^{2-}\} = 8 \cdot 10^{-10}$ mol/l. At pH values above 11, the carbonate activity will be of the same order of magnitude as the total dissolved inorganic carbon: $\{CO_3^{2-}\} \cong [CO_3^{2-}]_{tot}$.

Table 3.3.1

Distribution coefficients in concrete environments for $^{14}CO_3^{2-}$ reported in the literature.

K_D/m^3kg^{-1}	Reference
1 - 10	Bayliss et al., 1988
1.9 - 2.0	Hietanen et al., 1985
0.005 - 10	Andersson et al., 1982 and Allard et al., 1981
0.07 - 2.0	Hietanen et al., 1984a
0.01 - 0.15	Hietanen et al., 1984b

Table 3.3.2 reports calculated carbonate solubilities that have been obtained with the EQ3NR computer code (Wolery, 1983) using analyses of

cement pore waters reported in the literature. The important aspect of the results in Table 3.3.2 is that they indicate that the maximum solubility in a cement repository is around 0.23 GBq/l (=0.23 TBq/m^3=10^{-4} mol/l). A value of 23 MBq/l (=10^{-5} mol/l) is given by Pryke and Rees (1985).

This means that the amount of carbon-14 discussed here (5 TBq) might be dissolved in less than 25 m^3 of pore cement water. This would be so if all inorganic carbon in the water was carbon-14. However, one might expect that at encapsulation time the carbonate species in the pore water will be non-active.

Table 3.3.2

Carbonate solubility in cement pore waters (mol/l).

Chem Anal (mg/l)				Calculated solubility	
Na	Ca	pH	Ref	EQ3NR $[CO_3^{2-}]_{tot}$	Eqn 3.2.1 $\{CO_3^{2-}\}$
4.6	269.	12.4	a	$4.9\cdot10^{-6}$*	$5.0\cdot10^{-7}$
10.	88.	11.8	a	$6.4\cdot10^{-6}$**	$3.2\cdot10^{-8}$
100	480.	12.4	b	$4.9\cdot10^{-6}$	$5.0\cdot10^{-7}$
1500	90.	13.4	b, c	$3.7\cdot20^{-5}$	$5.0\cdot10^{-5}$

* Experimental value: $8.5\cdot10^{-5}$.
** Experimental value: $1.1\cdot10^{-4}$.

References: a: Bayliss et al., 1988
b: Andersson and Allard, 1986
c: Berner, 1987

As $^{14}CO_3^{2-}$ and $^{12}CO_3^{2-}$ are released from the ion exchange resins, the equilibrium of reaction 3.2.1 will be shifted to the right, and both active and non-active calcite will precipitate. Therefore the amounts of pore cement water needed to dissolve 5 TBq of ^{14}C are much higher than 25 m^3.

In order to use a solubility limit for inorganic carbon, the degradation time of concrete must be estimated. Neretnieks et al. (1987) have made calculations (in their Appendix A.5) on concrete degradation. If we use the approximate formulas of Atkinson and the Shrinking Core Model (Neretnieks et al., 1987), we find that a 0.8 m thick concrete wall is deteriorated in 1680 to 69 000 years, depending on the model used.

The best estimate for a degradation of 0.8 m thick concrete wall appears to be Eq A5.7 in Neretnieks et al. (1987), i.e., the Shrinking Core Model for sulfate intrusion from bentonite to concrete. This equation gives a penetration depth of 38.2 cm after 1000 y, and 80 cm degradation after 4370 y. The depletion of $Ca(OH)_2$ is calculated with Eq A5.6 in Neretnieks et al. (1987), giving 9.6 cm at 1000 y, and 80 cm at 69250 y. The results from Eqs A5.6 and A5.7 in Neretnieks et al. (1987), agree well with the results from CHEMTRN numerical calculations (Neretnieks et al., 1987, p A5-20).

These results are in agreement with other estimations (Atkinson, 1985) which are reviewed by Chapman and Flowers (1986). According to those calculations, the average pH in the near-field groundwater in a concrete repository is higher than 10.5 for more than 10^6 y.

We conclude that the large quantities of concrete in the Swedish SFR repository will probably reduce the mobility of ^{14}C for a time period of tens of thousands of years. Due to the simultaneous radioactive decay of ^{14}C, the retention of carbonate in the concrete matrix (as $CaCO_3(s)$), will reduce the amount of radiocarbon available for groundwater or gaseous transport. For example, if 5 000 GBq-^{14}C (= 221 g of calcite) are precipitated in the cement matrix at time t = 0, and due to a drastic change in the chemical conditions all the calcium carbonate is dissolved at t=50 000 y, the amount left after radioactive decay is only 11.8 GBq-^{14}C (= 0.52 g of $CaCO_3$ that can be dissolved).

Bacterial degradation of ion exchange resins and bitumen will produce some $CO_2(g)$ as discussed in section 3.2, and that might shorten the degradation time of concrete. The bacterially produced CO_2 will react with the alkaline components of cement (Dayal and Klein, 1988):

$$CO_2 + Na_2O \rightarrow 2Na^+ + CO_3^{2-}$$

$$CO_2 + Ca(OH)_2 \rightarrow CaCO_3 + H_2O$$

The buffer capacity of the repository will be determined by the amount of alkaline components present. If we use the data in Table 1.1, for the composition and amounts of concrete we get $\sim 1 \cdot 10^8$ mol $Ca(OH_2)$, $7 \cdot 10^5$ mol K_2O

and $2.8 \cdot 10^5$ mol Na_2O for the repository discussed here. The concrete composition listed in Table 1.1 is taken from Neretnieks et al. (1987, Appendix A1). Similar values are used by Lever and Rees (1988).

The microbial degradation of ion exchange resins and bitumen might produce $1.36 \cdot 10^8$ moles of $CO_2(g)$ within 10000 years as discussed in section 3.2. Thus, in principle the totality of $Ca(OH)_2$ in the repository might be neutralized during this period of time and the pH in the repository might decrease due to the chemical reaction, 3.4.1. However, some of the $CaCO_3$ precipitated will seal pores and fractures in the concrete, and the bulk of the cement will be protected from further attack. This is supported by the experimental observation that not all carbon in cement is available for isotope exchange (Bayliss et al., 1988). Nevertheless, the uncertainty in the rate of CO_2 microbial production is so high as compared with those of $CaCO_3$ precipitation/dissolution and CO_2-gas migration, that it is not possible to reach any quantitative conclusion.

The release of carbon-14 from a cement matrix has been studied by leaching experiments. Release of ^{14}C from carbon-14 carbonate inmobilized in cement has been studied by Burger (1979), Speranzini and Buckley (1981) and Scheele and Burger (1982). The leaching was increased mainly by high CO_2 contents of the leachate (Burger, 1979; Scheele and Burger, 1982). Leaching of ion exchange resins inmobilized in cement and bitumen matrix are given by Speranzini and Buckley (1981) and Habayeb (1985). Cement inmobilization appears to give the lowest release rates. Habayeb (1985) estimates 6.5% release after 500 years leaching of ^{14}C in Portland/epoxy resins.

3.4 Chemical reactions in the geosphere

As carbon is present in all natural waters, it is convenient to use radiocarbon to determine the apparent age of groundwater samples. For that purpose, the physico/chemical processes affecting ^{14}C in the geosphere have been extensively studied.

Groundwater ages are usually estimated with the carbon-14 method from analyses of inorganic carbon. Although ^{14}C might be also present in small quantities in dissolved organic compounds, this is usually neglected (Freeze and Cherry, 1979, p 135; Murphy et al., 1985). The main difficulty in ground water determinations is to estimate the amount of inactive carbon introduced in the groundwater since it became closed to the atmosphere.

In the soil zone, CO_2 is accumulated by plant respiration in the roots. Partial pressures for CO_2 of 10^{-2} atm are common. This carbon dioxide reacts with soil calcite or silicates (Fontes, 1983; Ross, 1988):

$$CO_2 + H_2O + CaCO_3 = 2HCO_3^- + Ca^{2+} \qquad \text{Eq 3.4.1}$$

$$CaAl_2Si_2O_2 + 8H_2O + 2CO_2 = Ca^{2+} + 2Al(OH)_3 + 2H_4SiO_4 + 2HCO_3^- \qquad \text{Eq 3.4.2}$$

It should be noted that if ^{14}C is present in the CO_2 gas, in the first of the above reactions, the ^{14}C activity of the dissolved HCO_3^- will be diluted by the carbon from the calcite, which usually is old and inactive.

Any other source or sink of CO_2 will also affect the ^{14}C activity in the groundwaters (Wigley et al., 1978) because carbon dioxide is usually in equilibrium with inactive calcite (Eqn 3.4.1). Common processes that affect carbon-14 through carbon dioxide production/consumption are: sulfate reduction and methane production (Pearson and Hanshaw, 1970; Barker et al., 1978; Wigley et al., 1978; Wetzel, 1983; Martens and Berner, 1977; Colombo et al., 1982):

$$CO_2 + 3H_2 \rightarrow CH_4 + H_2O$$

$$2(CH_2O) \rightarrow CH_4 + CO_2$$

$$O_2 + (CH_2O) \rightarrow HCO_3^- + H^+$$

$$SO_4^{2-} + 2(CH_2O) + 2H^+ \rightarrow 2CO_2 + H^+ + HS^- + 2H_2O$$

$$CH_4 + SO_4^{2-} \rightarrow HCO_3^- + H_2O + HS^-$$

Other inportant sinks of ^{14}C are:

- isotope exchange between aqueous bicarbonate and solid carbonates (usually calcite), which results in a dilution of the ^{14}C of the water (Fontes, 1983; Mozeto et al., 1984)
- carbonate precipitation/dissolution over the flowpath of a groundwater which reduces ^{14}C activity (Wigley, 1976).

Because of very slow rates, isotope exchange between organic and inorganic carbon in aqueous solutions should not be expected without microbial activity. For example, the following isotopic distribution rate between CO_2 and CH_4:

$$^{13}CH_4 + {}^{12}CO_2 \rightarrow {}^{12}CH_4 + {}^{13}CO_2$$

has a reaction halftime of 3 y at 600°C and 75 y at 500°C, and the value extrapolated to 300°C is over 10^6 y (Giggenbach et al., 1983).

When granitic groundwater reaches the repository discussed in this report, it will be at equilibrium (or near equilibrium) with the calcite present in fracture coatings of the granite. Due to the alkaline cement environment, pore water containing $^{14}CO_3^{2-}$ in the repository will also be near equilibrium with calcite. During mixing of groundwater with cement pore water, the mixture will probably become oversaturated with calcite (Liljenzin et al., 1986), some precipitation will occur (Eq 3.4.1) and part of the carbon-14 contents of the cement pore water will become fixed in the vicinity of the repository. The resulting mixture of cement pore water and granitic groundwater will travel further through the granitic rock matrix. The remaining carbon-14 will be affected by the geochemical processes discussed in this section, all of which will dilute carbon-14 activity.

Most of the calcite precipitated in the vicinity of the repository will contain inactive carbonate from the intruding groundwater. As there are about 10^8 mol $Ca(OH)_2$ in the repository, a maximum of $7.4 \cdot 10^9$ g of calcite could be formed around the repository. Of these, only 221 g might be $Ca^{14}CO_3$ (=5 TBq). If a sudden change in climatic or chemical/hydrological conditions should take place, such as to dissolve the calcite through reaction 3.4.1, the isotopic dilution would result in a specific activity of $2 \cdot 10^6$ Bq/kg-carbon in the groundwater. If radioactive decay is taken into account, as exemplified on p. 2:15, a much lower specific activity is obtained.

Carbon-14 methane leaving the repository will travel with the groundwater as long as the sulfate and oxygen concentrations are low, otherwise, sulfate reducing bacteria or methylotrophic organisms will consume methane to produce organic carbon compounds. Any excess of methane might be released to the atmosphere when the groundwater is discharged to the surface.

3.5 Dispersion mechanisms in the geosphere

Transport of solutes in fracture rocks is described by diffusion, advection (transport by the groundwater movement), dispersion (mixing occurring in flows through porous media), and sorption (ion exchange and solid surface reactions) (Grisak and Pickens, 1980; Neretnieks, 1981).

Literature studies on the retardation of radiocarbon in geologic media are summarized in Table 3.5.1.

Several analyses of the performance of repositories containing ^{14}C are also available in the literature. The parameters for ^{14}C sorption and diffusion selected in these studies are given in Table 3.5.2. This table also contains data used on studies of ^{14}C migration in aquifers (in relation to groundwater dating).

Diffusion coefficients (D_i, cm^2/s) for carbon-14 species in aqueous solutions are as follows (Moore, 1972; Neretnieks, 1985): $CO_3^{2-} = 1.8 \cdot 10^{-5}$, $HCO_3^- = 1.2 \cdot 10^{-5}$, $CH_3CO_2^- = 1.1 \cdot 10^{-5}$, $CH_4 = 4 \cdot 10^{-5}$. For larger organic anions, Neretnieks (1985) gives $D_i < 10^{-5}$ cm^2/s.

The rock matrix acts as a reservoir for C-14 by diffusion from groundwater in the fractures. This might be an important mechanism for carbon-14 retardation (Fritz, 1983; Sudicky and Frind, 1981). This process is exemplified in Section 4.

Table 3.5.1

Experimental studies on the retardation of ^{14}C in geologic media.

Reference	Comments
Reardon et al., 1986	K_D= 0.15 - 0.3 ml/g (in soil)
Andersson et al., 1982 & Allard et al., 1982	Sorption in Ca-minerals K_D = 0.8 - 83 ml/g
Garnier, 1985	$D_L = \frac{u \cdot l}{Be}$ (in Table 2) = $(0.8 - 4) \cdot 10^{-9}$ m^2/s
Burkholder et al., 1976	Sorption = 0.1 (nuclide velocity/water velocity) for desert soil
Lang et al., 1986	D = $5.2 \cdot 10^{-7}$ cm^2/s (littographic rock) R_s = 3 - 20 ml/g " "
Mozeto et al., 1983	Isotope exchange between aqueous carbonate-calcite
Münnich et al., 1967	Ibid
Wendt et al., 1967	Ibid
Hietanen et al., 1985	Sorption in concrete-sand aggregate K_D = 1 - 3 ml/g
Muurinen et al., 1986	Diffusion in crushed rock and bentonite, D = $6 \cdot 10^{-11}$ - $4 \cdot 10^{-12}$ m^2/s
Pinnoja et al., 1984	K_a = $(<<1 \text{ to } 30) \cdot 10^{-4}$ m^3/m^2
Pinnoja et al., 1986	K_D $7 \cdot 10^{-4}$ - $6 \cdot 10^{-2}$ m^3/kg

Table 3.5.2

Dispersion and sorption parameters for ^{14}C in the literature.

Reference	Comments
Garnier, 1987	$D_o = 0.67 \cdot 10^{-9}$ m²/s $D_L = 528$ m²/y $= \frac{u \cdot L}{Be}$ with Be = 200 or 20
Sudicky and Frind, 1981	Dispersivity 10 m $D^o = 10^{-5} - 10^{-8}$ cm²/s
Pinner and Maple, 1986	$D_e = 3 \cdot 10^{-8}$ m²/t (concrete) $K_D = 0$ (mudstone) $= 2$ (clay) m³/t Dispersive length: $X_L = 1 - 10$ m $X_T = 0.1 - 0.5$ m Diffusion coefficient (m²/y): $D_{L,T} = 0.04 - 0.1$ (trench) $D_L = 6 \cdot 10^{-4} - 0.04$ (clay, mudstone)
Smith et al., 1987	Dispersion coefficient (m²/y): D = 0.1 (shale) or 0.003 (clay) Sorption = 8 (water velocity/nuclide velocity) Diffusion coefficient = $4 \cdot 10^{-6}$ m²/y (concrete)
Bush et al., 1984	Dispersion coefficient = 0.003 m³/d
Andrews and Pearson, 1984	$K_D = 0$ in calcium-free sandstone
Viemo et al., 1987	K_D $5 \cdot 10^{-4} - 10^{-3}$ m³/kg (crushed rock)

4 Numerical simulation of ^{14}C transport from a granitic repository

The most probable exposure pathways to humans from ^{14}C disposed in a granitic repository, are through groundwater or gas transport in the fracture network of the bedrock.

Previous sections have given an overview of the processes involved in the transformations of radiocarbon and its migration.

In this section we report the results of calculations for a simplified scenario. The assumptions and parameters used are descibed in section 4.2.

Other numerical simulations of radionuclide transport in the rock matrix have already been made for the SFR repository at Forsmark (Neretnieks et al., 1987; Neretnieks et al., 1988). Models for contaminant transport in fractured media are still subject to continuous research (see, for example, the recent papers by Berkowitz et al., 1988 and Fogden et al., 1988). The modelling exercise presented here concerns only transport of carbon-14, and our purpose is to give a description of the dispersion of this radionuclide in the rock matrix.

4.1 The computer program

We have used GEOPATH, a computer program that calculates transport of radionuclides in fractured and porous media (Marklund and Kjellbert, 1980).

GEOPATH is a two-dimensional algorithm based on finite differences, that takes into account processes like mass balance (for finite rectangles), nuclide decay, sorption and transport.

4.2 Parameters and assumptions

We have used a scenario that consists of the repository presented in Table 1.1. We have assumed that no bacterial activity takes place in the repository (i.e., no gaseous release of ^{14}C although the diffusion coefficients for the different carbon species, including methane, do not differ considerably). We have also assumed that the repository remains intact for 2500 y, and at that time, the repository is severely fractured, and ^{14}C is carried away by groundwater circulating through it. At 2500 years after repository closure, the part of the Scandinavian peninsula surrounding the repository at Forsmark has been elevated above the sea surface.

The repository is surrounded by a porous medium (a bentonite clay/sand layer 1.2 m thick on the sides, and 2.5 m on the top and bottom). On top of the repository there is a void space filled with crushed rock (size: 10x32.4x23.6 m), on top of the "void" space there is 40 m of granitic rock to the rock surface (i.e. the biosphere, the soil layer being neglected). A fracture, with an aperture of 10^{-3} m, is supposed to conduct water from the "void" space at the top of the repository to a primary recipient (discharge zone).

The groundwater flow is assumed to be horizontal, and the primary recipient for the biosphere (well, lake etc) is assumed to be 160 m downstream. This can be compared with calculated path lengths between 90 and 200 m for particles traveling between the repository and the rock surface with an upward directed groundwater flow (Carlsson et al., 1987). At time=2500 years, the flow pattern is assumed to be horizontal, and therefore the path lengths should be longer.

The groundwater in the vicinity of the repository is assumed to have a higher groundwater flow. This is due to the disturbance that the repository introduces in the natural groundwater flow. The thickness of the disturbed rock is assumed to be 10 m.

Table 4.2.1 presents the hydrodynamic dispersion coefficients and average pore water velocities used in the GEOPATH calculations. Figure 4.2.1 shows in a schematic diagram the repository surrounded by the bentonite/sand buffer and the fracture in the rock matrix.

The pore water velocities in Table 4.2.1 were calculated from the porosities and water fluxes given in Table 1.1. The amount of water that is discharged from the rock matrix surface (145x23.6 m^2) of the primary recipient of the biosphere is 17.2 m^3/y, while 1.64 m^3/y are delivered from the fracture.

The dispersion coefficients include hydrodynamic dispersion and ionic (or molecular) diffusion. We have not included sorption in our calculations.

Table 4.2.1

Hydrodynamic dispersion coefficients and average pore water velocities used in the GEOPATH calculations.

Material	Dispersion coeff. Horizontal m^2/s	Dispersion coeff. Vertical m^2/s	Pore water vel. Horizontal m/s	Pore water vel. Vertical* m/s
Repository	$1 \cdot 10^{-12}$	$1 \cdot 10^{-12}$	0.	$-1.6 \cdot 10^{-9}$
Repository wall (degraded concrete)	$1 \cdot 10^{-12}$	$1 \cdot 10^{-12}$	0.	$-1.6 \cdot 10^{-9}$
Bentonite	$1 \cdot 10^{-12}$	$1 \cdot 10^{-12}$	0.	$-1.6 \cdot 10^{-9}$
Granite matrix	$1.6 \cdot 10^{-6}$	$5 \cdot 10^{-14}$	$1.6 \cdot 10^{-6}$	0.
Void space filled with crushed rock	$1.6 \cdot 10^{-10}$	$5.3 \cdot 10^{-12}$	$1.6 \cdot 10^{-9}$	$-5.3 \cdot 10^{-11}$
Granite near the repository	$3.2 \cdot 10^{-6}$	$5 \cdot 10^{-14}$	$3.2 \cdot 10^{-6}$	0.
Fracture	$2.2 \cdot 10^{-7}$	$1.0 \cdot 10^{-9}$	$2.2 \cdot 10^{-6}$	0.

* Negative values indicate upward flow

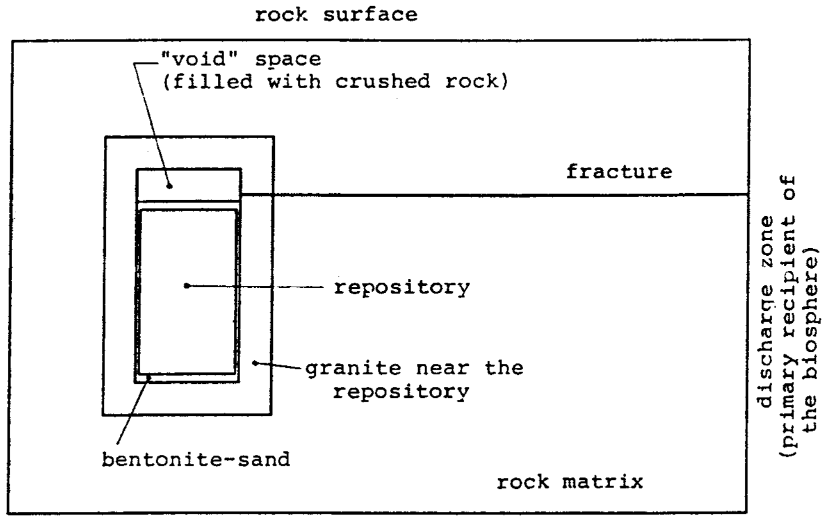

Figure 4.2.1

Schematic view of the model repository situated in a granitic rock matrix. The scenario is 232 m wide and 145 m high. The groundwater flow in the granitic rock is directed towards the right.

Diffusivities in the range $1\text{-}200 \cdot 10^{-12}$ m^2/s are given for several aqueous solutes in wet bentonite by Neretnieks (1984). Diffusivities in granite are given by Skagius and Neretnieks (1986), Jefferies (1987), and Birgersson and Neretnieks (1988). The values for diffusivities given by these authors are in the approximate range $1\text{-}50000 \cdot 10^{-14}$ m^2/s.

Dispersivities for transport in granite are given by Goblet et al. (1979) and Calmels et al. (1979) in the range 0.6 to 4 m. Abelin et al. (1985) give Peclet numbers in the range 0.9-37, which with a dispersion length of 5 m would give dispersivities in the range 0.14 to 5.6 m. We have arbitrarly selected a value of 1m for the dispersivity in the granite, and 0.1 m in the void space (above the repository) filled with crushed rock. The longitudinal dispersivity for the fracture was taken equal to 0.1 m.

The initial concentration of carbon-14 (at time = 2500 y) in the repository and concrete walls is set equal to 0.14 GBq/m^3. This gives a total inventory of 5TBq. With a porosity of 0.2 m^3/m^3, the concentration of carbon-14 in the pore water of the repository is then assumed to be $3 \cdot 10^{-7}$ mol/l, which is below the solubility limit for cement pore water discussed in section 3.3.

In reality, however, the initial concentration will be lower. When ^{14}C-carbonate is released from the ion-exchange resins, calcium carbonate will become oversaturated and precipitation will occur, probably at the surface or near vicinity of the ion-exchange particles.

4.3 Results

The calculations were made for the time interval 2500 to 102500 years, and the results are reported in Figure 4.3.1. The total amount of carbon-14 discharged to the primary recipient is 8.4 GBq. The contribution from the fracture is small (8.9 MBq). The maximum discharge rate is $4.5 \cdot 10^5$ Bq/y.

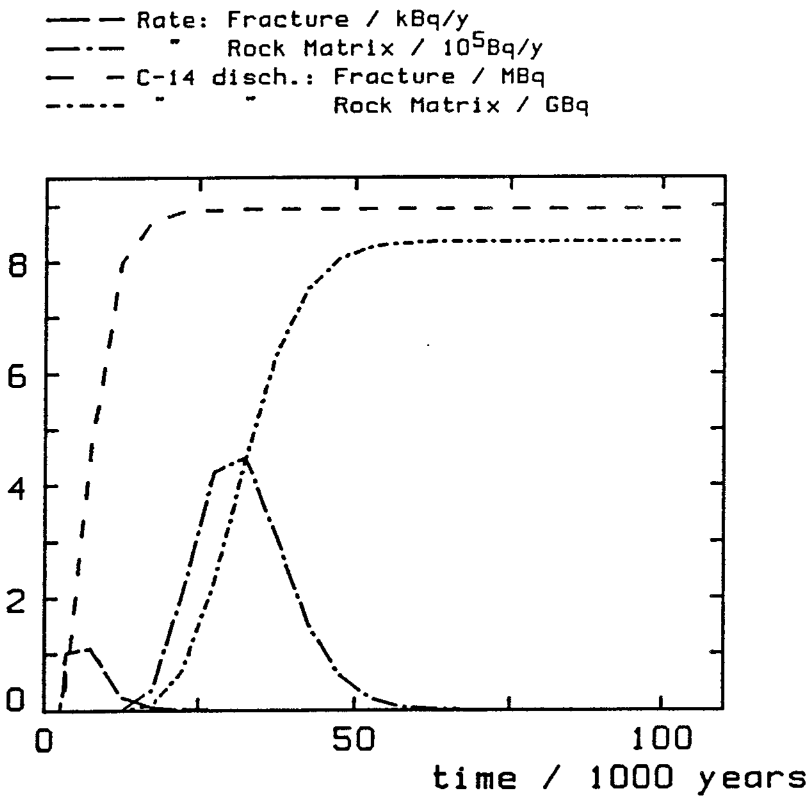

Figure 4.3.1

Results from the GEOPATH calculations on the migration of carbon-14 in the geosphere. The discharge rate to the primary recipient has maximum values of $1.1 \cdot 10^3$ and $4.5 \cdot 10^5$ Bq/y for the fracture and rock matrix respectively. The accumulated discharge is 8.9 MBq for the fracture and 8.4 GBq for the rock matrix.

The results of the GEOPATH calculations are also shown in Figure 4.3.2 at times 7500 and 32500 years. These time intervals correspond to the initial time, as well as to the maximum release rates for the fracture and the rock matrix. The vertical axis in Figure 4.3.2 is proportional to the concentration of ^{14}C (in Bq/m^3), but the figures have been rescaled to an arbitrary height, and to a logarithmic scale which enhances the areas of the scenario with low concentrations of carbon-14.

The values in Figures 4.3.1 and 4.3.2 should be taken only as an illustration of the transport processes involved. A quantitative discussion should include a sensitivity analysis on the input parameters, because some of them contain assumed values which should be discussed more thoroughly. However, a quantitative discussion of the transport processes in the geosphere is outside the scope of this report. The reader is referred to the work of Bailey and Marine (1980), for example.

Nevertheless, the calculation shows that most of the carbon-14 leaves the repository at an early stage, in agreement with the results of Wiborgh et al. (1987b). Figure 4.3.2 shows that most of the carbon-14 remains in the bentonite/sand and crushed rock layers above the repository. Geosphere dispersion, which is qualitatively illustrated with this calculation, can decrease the total release of carbon-14 by some orders of magnitude. Similar results are reported by Bush et al. (1984) who compare doses calculated from release of ^{14}C to the atmosphere, aquatic environments or to the geosphere (Figure 8.9, Bush et al., 1984).

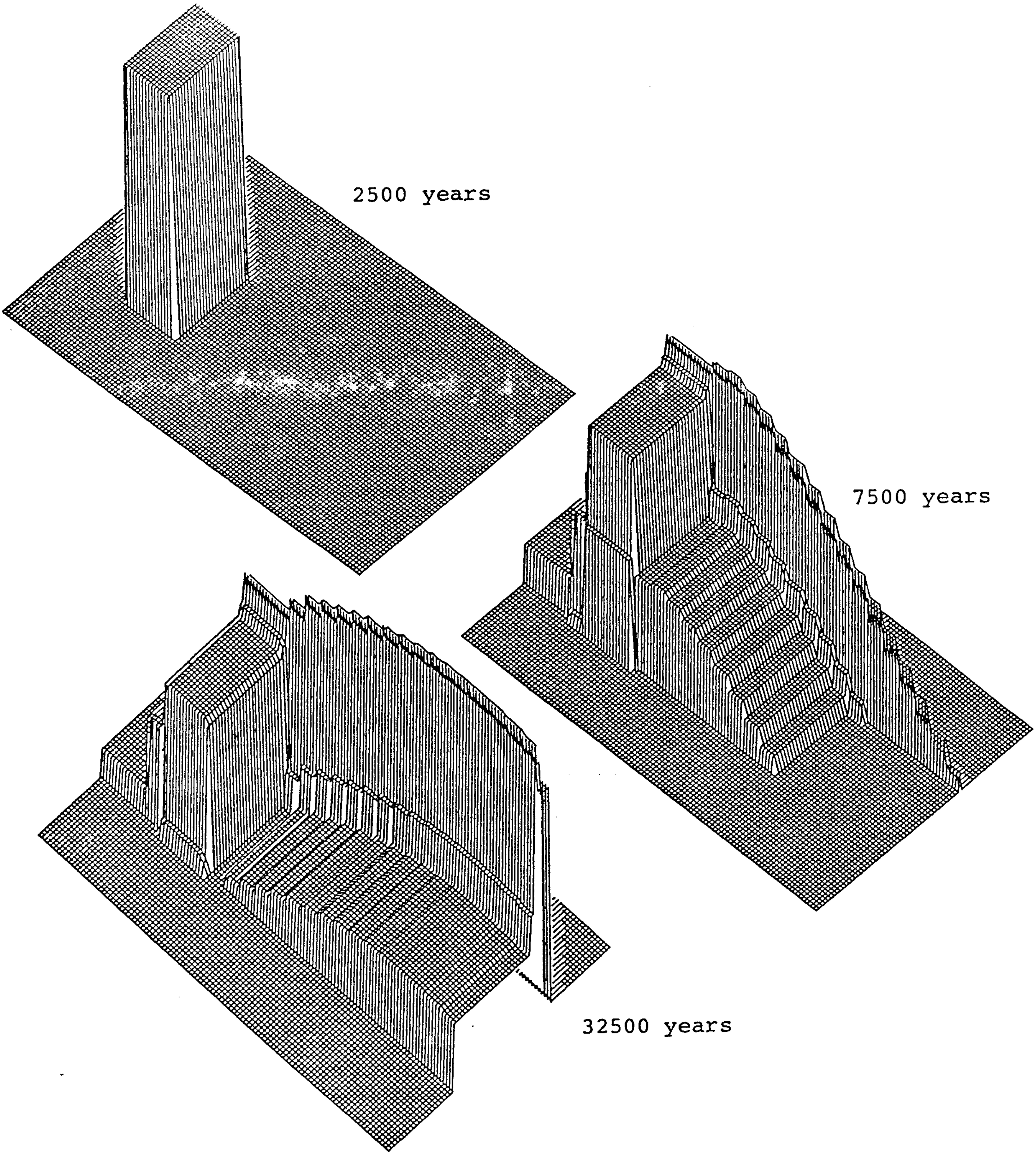

Figure 4.3.2

Results from the GEOPATH calculations on the migration of carbon-14 in the geosphere. The diagrams show the concentration of ^{14}C (Bq/m^3) s the vertical height (the Z-axis). An arbitrary scale is used in the Z-axis, as the concentrations decrease exponentially with time. Each step in the X/Y plane is equivalent to 2 m. The discharge zone is at the lower right edge of each figure.

5 Conclusions

In this report we try to describe the processes that may affect carbon-14 both in a granitic repository and on its way to the biosphere. A literature survey has been made, and some simple calculations illustrate the most important phenomena. The main conclusions are:

- carbon-14 is expected to be present in low and intermediate level nuclear waste repositories as $^{14}CO_3^{2-}$ species in anion exchange resins. However, there is some uncertainty in the carbon-14 contents of the waste.
- the average pH values of the pore water in a concrete repository are expected to remain ≧10.5 for a period of at least 10^6 years.
- total inorganic carbon concentration of the concrete pore water is expected to be controlled by $CaCO_3$ saturation. Addition of carbonate, either by release of $^{14}CO_3^{2-}$ (and CO_3^{2-}) from the ion-exchange resins, or by intrusion of granitic groundwater, will cause the precipitation of calcium carbonate. In this way most of the 134 g of $^{14}CO_3^{2-}$ (=5 TBq) initially present will be retained in the vicinity of the repository by reaction with some of the ~10^8 mol $Ca(OH)_2$ that are initially present in the cement matrix of the repository
- bacterial production of CO_2 and CH_4 from the decay of ion-exchange resins and bitumen might contribute to ^{14}C release. However, the information available in the literature on the rate of such processes is scarce and a large uncertainty in the release of ^{14}C is associated with microbial reactions. For example, high pH values (≧10.5) expected in a cement repository might reduce the rate of microbial gas production by some orders of magnitude.
- hydrodynamic dispersion of carbon-14 will greatly reduce its discharge to the primary recipient, even when no sorption is taken into account.

Long-term radiological exposure to Carbon-14 from a final repository for reactor waste (SFR)

1 Introduction

The Swedish repository for low- and intermediate level wastes (SFR) is situated under the Baltic Sea, at a depth of at least 50 m below the rock surface (SFR 1, 1987). The individual and collective doses resulting from long-term releases of radionuclides from the SFR installations were assessed by Bergström and Puigdomenech (1987). Two scenarios were treated:

- release to a coastal area of the Baltic Sea, starting immediately after repository closure
- release to an inland area in which a well and a lake were situated, starting 2500 years after repository closure

Due to the present steady rising of the Scandinavian peninsula, the coastal area of the Baltic Sea might in time be transformed into inland, which justifies the second approach. The chosen starting time for radionuclide release - 2500 years after repository closure - is the earliest time when a lake might form within that part of the area.

According to the results by Bergström and Puigdomenech (1987), the inland scenario gave rise to the highest doses, and C-14 constituted one of the main dose contributors. The exposure from C-14 was dominated by consumption of fish from the lake, followed by water consumption from the well. C-14 was assumed to be completely available to biological uptake which had important consequences on the calculated doses from fish consumption.

The aim of this part of the report is to present a closer analysis of the carbon flow in the brackish and fresh water ecosystems which may constitute the future carrier for C-14 discharging from the SFR repository, and to assess the individual and collective doses due to C-14 exposure. The amounts and distribution of carbon species in different aquatic environments are reviewed. Carbon fluxes both within a number of freshwater lakes, and between the lakes and the surrounding biospheric compartments, are derived from published data. The model structure and attendant input data are presented. The results from computer simulations on carbon releases from the SFR repository are compared with data on the specific activity of carbon in the different biospheric compartments. The calculated global dose commitment is compared with those given by other authors in order to validate the model results.

Ragnar Gelin wrote Chapter 5 dealing with dose conversion factors for ^{14}C, and Ignasi Puigdomenech made the computer simulations. The advice and critisism of Ulla Bergström and Ignasi Puigdomenech, and the considerate assistance of Elisabet Appelgren are gratefully acknowledged.

2 Carbon in aquatic environments

Most carbon in marine and fresh waters occurs as equilibrium products of carbonic acid (H_2CO_3). A smaller amount of carbon occurs in organic compounds as particulate and dissolved detrital carbon, and a minor fraction occurs as carbon incorporated in living biota. The carbon content of the water is dependent upon the equilibrium established between atmospheric CO_2, the bicarbonate-carbonate system, and upon contributions from metabolic respiration and utilization in photosynthesis. Carbon contents and major constituent composition of natural waters are given in Table 1. The different fractions of carbon making up the total carbon content in marine and fresh waters, respectively, are viewed in Figure 1.

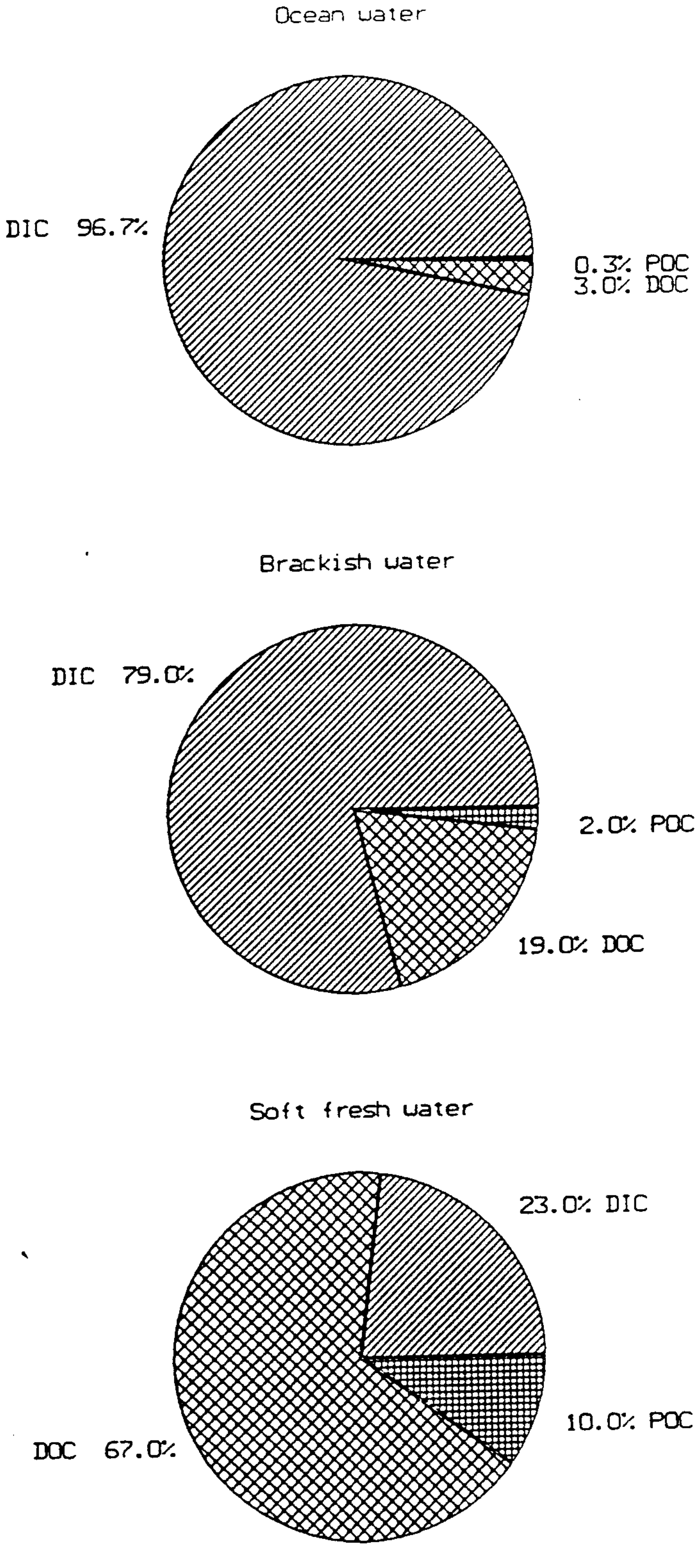

Figure 1

Carbon species composition in natural waters.

Table 1

Carbon contents and chemical composition of natural waters.

All values in mg/l (ppm)

HCO_3^-	C	Cl^-	SO_4^{2-}	Na^+	K^+	Ca^{2+}	Mg^{2+}	Ref	Location/Descript.
Sea water:									
142	28	193500	2710	10760	399	411	1290	A	
	0.5 (organic C)							B	
Baltic Sea:									
85	16.7	3100	490	1900	63	95	220	C	at Håtö 9m depth
Average stream:									
58.4	11.5	7.8	11.2	6.3	2.3	15	4.1	D	
Rain water:									
		0.6	3.2	0.4	0.3	0.9	0.2	E	Stockholm's rain
Scandinavian Lakes:									
6.4	1.3	11.2	2	7	0.2	1.1	0.9	F	Salsvatn, 300m
1130	222	16000	1920	8910	384	384	1050	F	" 410m
69	13.6	1538	241	850	32	216	--	G	Sibbo 6m
30	5.9	28	39	20	3.6	25	--	G	Trobbo
70	13.8	1200	190	630	27	190	--	G	Strömst
26	5.1	4	9	3.5	1.4	12	--	G	Hässle
Brackish Swedish wells:									
223	44	1750	256	-	-	509	-	D	Håtö 7, 39m depth
53	10	1600	<1	523	12	386	4	D	Wilh.-lund 2, 139m
262	52	690	86	378	5.4	100	16	D	Hässelby 1, 100m
931	183	2500	120	1527	95	57	76	D	Åker 11, 28m
Swedish groundwaters:									
									Forsmark-SFR:
77	15.2	4200	490	1500	16	1600	170	H	HK1 43-62m
48	9.4	4000	150	1300	6.2	1000	100	H	HK2 81-116m
53	10.4	2700	130	760	10	1100	130	H	HK3 81-101m
96	18.9	5000	480	1100	44	760	180	H	HK7, P1
98	19.3	5200	670	1600	19	1400	250	H	HK10, P1
81	16	4200	560	1700	30	970	190	H	HK11, P1
									Finnsjön
393	77	75	24	170	2.8	22	41	I	Fi4, 534m
41	8	5600	320	1500	8	1800	100	I	Fi5, 384m
									Klipperås
134	26	18	<.5	27	1.1	30	1	J	Kl2, 326m
317	62	25	41	69	4.7	56	13	E	Kopparmora, Värmdö
63	12	5000	300	1200	21	1470	266	E	Byholmen "
123	24	4790	792	1690	25	1026	226	E	Fruvik "
15	3	211	1.4	710	5.9	570	24	E	Småris "

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2.1 Inorganic carbon

Both oceanic and fresh waters are close to equilibrium with atmospheric CO_2. In the marine habitat, the dissolved inorganic carbon (DIC) pool (DIC = CO_2^- + HCO_3^{2-} + CO_3) contains about 2 mmol C l^{-1} (28 ppm), largely as HCO_3^-. In the Baltic Sea, the DIC content is about 17 ppm (Table 1). In fresh waters, the total inorganic carbon content is much more variable, within a typical range of 50 µmol to 10 mmol l^{-1} (0.6 - 120 ppm). The eutrophic Lake Trobbofjärden, middle Sweden, contains 5.9 ppm while the adjacent Lake Sibbofjärden, which has a narrow connection with the Baltic Sea, shows a DIC content close to 14 ppm. The proportions of CO_2, HCO_3^- and CO_3^{2-} at various pH values in fresh water are given in Figure 2 and Table 2. Free CO_2 dominates in water at pH 5 and below, while above pH 9.5 CO_3^{2-} is quantitatively significant. Within the normal range of pH, range of pH, HCO_3^- predominates.

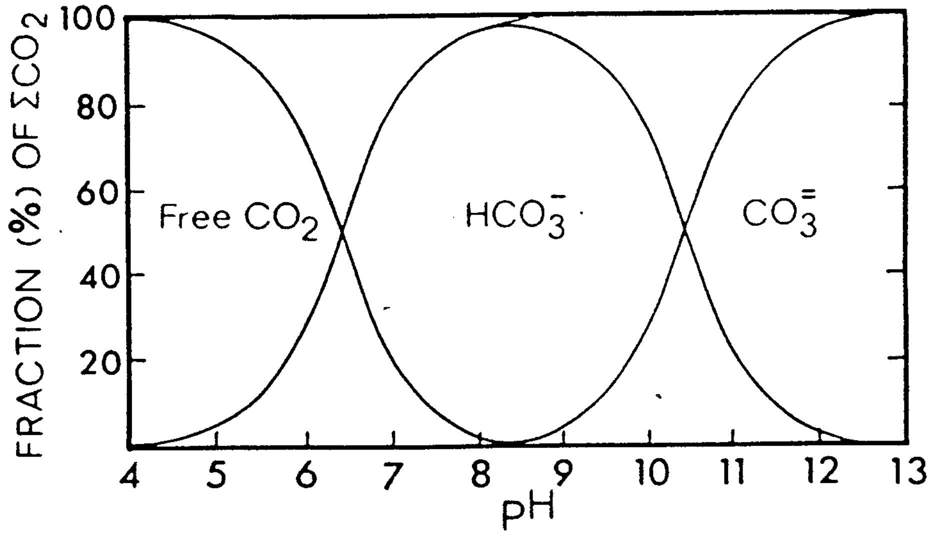

Figure 2

Relation between pH and the relative proportions of inorganic carbon species in solution at various pH values.

Table 2

Proportions of inorganic carbon species in water at various pH.

pH	TOTAL FREE CO_2	HCO_3^-	$CO_3^=$
4	0.996	0.004	1.25×10^{-9}
5	0.962	0.038	1.20×10^{-7}
6	0.725	0.275	0.91×10^{-5}
7	0.208	0.792	2.6×10^{-4}
8	0.025	0.972	3.2×10^{-3}
9	0.003	0.966	0.031
10	0.0002	0.757	0.243

2.1.1 CO_2 exchange between the atmosphere and the water

The flux of carbon between the atmosphere and ocean water is largely determined by the carbonate-bicarbonate equilibrium in the surface waters, but is also affected by mixing and circulation that remove carbon from the surface waters into deeper layers where it may be isolated for some time. The residence time for CO_2 in the well-mixed surface layer of the sea is less than 10 years, while for the deep waters it is about 1000 years (Stuiver, 1973). The transfer rate of CO_2 between the atmosphere and the surface ocean can be estimated, based on the assumption that the net invasion of naturally produced CO_2 into ocean water is fast enough to balance the rate of decay of ^{14}C residing in the sea. From knowledge of the entire ocean ^{14}C inventory and of the difference between the $^{14}C/C$ ratio of atmospheric CO_2 and of ocean water, the gas exchange rate can be calculated. The estimated flux from atmosphere to ocean water was 19±6 moles of CO_2 $m^{-2}y^{-1}$ or 228±72 g C $m^{-2}y^{-1}$ (Broecker et al., 1979). A second approach is based on the deficiency of radon gas observed in the surface ocean mixed layer (Broecker and Peng, 1974). The best radon based estimate for the CO_2 exchange rate was 16 moles $m^{-2}y^{-1}$ or 192 g $m^{-2}y^{-1}$ (Peng et al., 1979), a value also adopted by Oeschger et al. (1975) and Broecker et al. (1979) in their model calculations. Likewise, in a global carbon model developed by Nair (1981), an atmosphere-to-surface ocean flux of 340 g $m^{-2}y^{-1}$ was calculated for "cold surface oceans". The reverse flux from ocean surface-to-atmosphere was estimated to be 210 g C $m^{-2}y^{-1}$.

In the limnic environment, many lakes with pH near neutrality are slightly supersaturated with CO_2 relative to the atmospheric pressure of CO_2. However, the exchange of CO_2 between atmosphere and water cannot be determined by partial pressure differences alone. A flux of atmospheric CO_2 may occur into poorly buffered waters of low DIC content. In a Canadian soft-water lake, atmospheric invasion of CO_2 amounted to 0.12 ± 0.06 g C $m^{-2}d^{-1}$, accounting for 30 - 90 % of the carbon fixed by phytoplankton (Schindler et al., 1972). In more productive waters, consumption of CO_2 by photosynthetic activity enhances the flux of atmospheric CO_2 to the water (Emerson, 1975). At the opposite extreme, in a hard-water lake that received high inputs of inorganic carbon, CO_2 was released into the atmosphere throughout the year (Otsuki and Wetzel, 1974). An average exchange rate of 50 g C $m^{-2}y^{-1}$ was assumed to occur in both directions through the air-water interface in Canadian lakes (McKee and Rowsell, 1984).

2.1.2 Utilization of inorganic carbon by algae and macrophytes

Inorganic carbon is a major nutrient of photosynthetic metabolism. Free CO_2 is most readily utilized by nearly all algae and larger aquatic plants (Allen and Spencer, 1981) and photosynthesis and respiration are two major factors that influence the amounts of CO_2 in water. Besides, the primary producers also require nitrogen, phosphorus and a wide range of trace elements. Phosphorus and nitrogen limit photosynthesis more frequently than does inorganic carbon, which occurs in much greater abundance.

By photosynthesis CO_2 is converted to organic matter and O_2. The composition of organic matter in plankton is approximately $C_{106}H_{263}O_{110}N_{16}P_1$ (Redfield et al., 1963). Thus, photosynthesis can be represented by the equation

$$106CO_2 + 16NO_3^- + HPO_4^{2-} + 122H_2O + 18H^+ \text{ (+ trace elements, energy)} \rightleftarrows C_{106}H_{263}O_{110}N_{16}P_1 + 138O_2$$

This illustrates the enormous importance of phosphate in the carbon-oxygen balance of waters. Each atom of P added to a surface water results in the fixation of about 106 atoms of carbon in organic matter, and when the organic matter produced from one atom of P decays, it has the potential to consume 138 molecules of oxygen. The annual productivity of temperate-zone lakes is thus primarily a function of the phosphorus supply; lakes with exceptionally high P loadings may be temporarily limited by nitrogen or carbon, with the upper limit determined by the light supply (Schindler, 1976).

The maximum phytoplankton uptake of DIC in a U.S. subalpine lake represented less than 1 % of the amount of DIC present in the photic zone (Rau, 1978). Plankton respiration was also small, indicating that water-column photosynthesis and respiration processes must have little influence on the DIC concentrations. A considerable buildup of DIC occurred during the winter. Dissolved oxygen concentrations in deep water were inversely related to DIC concentrations, indicating that benthic respiration was generating some of the inorganic carbon present. Sedimentation studies showed that terrestrial detritus must contribute most to the organic carbon pool of the benthos. Annual benthic respiration exceeded the sedimentation of lake-produced organic carbon by a factor of 3 (Wissmar et al., 1977), thus underlining the metabolism of allochthonous substrates as a source of DIC. In conclusion, much of the DIC available for phytoplankton production seems to be previously reduced and oxidized. Nonpelagic sites must participate in both the fixation and respiration processes that precede carbon refixation in the lake.

2.2 Organic carbon

The organic matter of soils and waters can be viewed as a mixture of plant and animal products in various stages of decomposition. This complex system may be simplified by separation into two categories: nonhumic and humic substances. Nonhumic substances include carbohydrates, proteins, amino acids, fats and other low-molecular-weight organic substances which generally are relatively easily utilized and degraded by microorganisms, and which exhibit rapid flux rates. Because of rapid rates of utilization, the concentrations of nonhumic substances in water are usually very low. Humic substances form most of the organic matter of soils and waters, and include acidic compounds of high molecular weights which are relatively resistant in aquatic systems.

Allochthonous sources of organic matter to aquatic systems are primarily of terrestrial plant origin. The input results from direct leaching from living vegetation or from soluble compounds carried by runoff from dead plant material in various stages of decomposition. The particulate organic matter (POM) entering lake waters is leached of a significant part of its organic content as dissolved compounds. The rates of decomposition of dissolved organic matter (DOM) are rapid (days) in comparison with POM (weeks as regards leaves, years for woody material). Approximately 10 times more organic matter occurs in the dissolved phase than occurs as POM. However, as the total organic matter of lake water increases, the percentage in the dissolved fraction increases disproportionally to that of the particulate fraction.

Most lakes are small and the littoral zone constitutes a high proportion of the surface area. That implies that the littoral together with allochthonous sources form the major inputs of organic carbon to the lakes. Autochtonous sources of POM and DOM include degradation of macrophytes and attached flora, and of primary producers of the pelagic zone.

2.2.1 Dissolved and particulate organic carbon

The range of total organic carbon in natural waters is 1 - 30 ppm. Almost all of the organic carbon consists of dissolved organic carbon (DOC) and dead particulate organic carbon (POC). Production of DOC and POC is a result of autotrophic and heterotrophic activity, both in the lake and within the surrounding drainage area. For practical reasons, POC is defined as the carbon fraction that is retained on a 5 µ filter. The two fractions are thus merely an arbitrary division of a smooth continuum of large particles to small molecules. Hence, DOC concentrations frequently include a significant collodial fraction to be added to the truly dissolved organic carbon, especially in heavily stained waters.

The major pool of organic carbon is in the dissolved form; the ratio of DOC to POC often approximates 6:1 to 10:1 in fresh water systems (Wetzel and Rich, 1973). World DOC average of rivers is 5.8 ppm (Meybeck, 1982). The origin of DOC in aquatic ecosystems are largely photosynthetic. Organic carbon is autotrophically synthesized within the water body by the pelagic and littoral flora and DOC is added to the pool through secretion and autolysis of cellular contents. Furthermore, allochthonous DOC is generated in the terrestrial systems of the drainage area and transported by runoff to the recipient basin. Much of the DOC originating from terrestrial and marsh plants has humic characteristics and is resistant to rapid bacterial degradation. The allochthonous input of DOC from the drainage basin and the export from the lake represent a major flow of carbon through the lake.

Autochthonous primary production by the pelagial and the littoral flora are the two major contributors to the POC of natural lake systems. Allochthonous POC is relatively small in contrast to the major inputs of DOC.

The content of DOC in the Baltic Sea ranges from 1 to 13 ppm, with an average of about 4 (Kay, 1954; Skopintsev et al., 1959; Szekielda, 1968, 1971; Ehrhardt, 1969; Fonselius, 1972; Jurkovskis and Luke, 1974; Pecherzewski and Lawacz, 1975).

In the fully marine environment, the DOC content is generally less than 3 % of the total content, or about 1 ppm (Bolin et al., 1981).

2.2.2 Distribution of organic carbon

A large fraction of the organic carbon consists of refractory compounds which are chemically stable, of low stability, and resistant to microbial degradation. The remaining fraction consists of labile compounds which are readily utilizable and which cycle rapidly, representing the major pathways of carbon fluxes. In lakes in general, detrital organic matter is the dominant post of organic carbon. Detritus consists of dead organic carbon lost from all trophic levels or from external sources of the ecosystem, and is discernible from living organic carbon and from inorganic carbon. Much of the autochthonous production of detrital matter originates from benthic littoral and wetland vegetation and forms an excess carbon reserve that is refractory to rapid metabolism. The relatively slow utilization rate of this detrital reservoir gives stability to aquatic systems. Detrital DOC and POC have long been known to exceed many times the amount of organic carbon present as living material in the form of bacteria, plankton, flora and fauna (Saunders, 1969; Wetzel et al., 1972). The amount of carbon in higher organisms is trivial in comparison with the total amount of carbon in the lake, and the trophic structure above the producer-decomposer level, with all its complex interactions, has a relatively minor impact on the total carbon flux of the system.

Rapid transformations by microbial activity progressively degrade organic matter to CO_2 and heat. However, the amount of organic carbon utilized and transformed by animals is a quantitatively small portion of that of the whole system; frequently more than 90 % of the organic matter entering the system from photosynthetic sources is not utilized in the grazer and detrital food chains. Detrital metabolism occurs principally in the benthic region, where a majority of POC is decomposed, and the sediment becomes the major place of transformation in many lakes.

2.3 Carbon in sediments

In most lakes, organic carbon is a mixture of compounds derived from terrestrial, littoral, planktonic and bacterial sources. Cellulose and similar materials make up much of the organic matter that enters the sediments. The organic carbon in superficial sediments of the Great Lakes (USA) comprised 65 - 85 % humic compounds, 10 - 12 % amino acids, 2 - 6 % carbohydrates, 2 - 8 % lipids and 0.4 - 4 % amino sugars (Kemp and Johnson, 1979); total organic carbon (TOC) concentrations ranged from 0.5 to 5 % dry wt (Callender, 1969; Kemp and Lewis, 1968). In Lake Superior, TOC concentrations averaged 3 - 4 % (Johnson and Evans, 1982) and most cores showed a pronounced decrease in TOC content with burial depth. A mean concentration of 2.1 - 3.7 % was recorded in sediments less than 100 years old. The average content of TOC in profundal sediments from 16 Canadian lakes was 20.4 ± 7 % with a range of 8 - 34 % (Brunskill et al., 1971). The relatively high percentage of TOC was due to an abundant supply of terrestrial organic matter to the sediments. For a number of lakes, sediment carbon was found to be directly related to the theoretical water renewal time:

$$C = 32.17 - 0.97\ T$$

where

C = carbon content (% dry wt)
T = water renewal time (y)

The factors controlling sedimentary TOC in the freshwater environment, e.g. productivity, water depth and sedimentation rate, are also the primary factors affecting sedimentary TOC in the marine environment.

Organic carbon in most marine sediments is primarily autochthonous. The carbon content of superficial (0 - 4 cm) sediments collected at 7 m depth in the shallow Narragansett Bay, Rhode Island, USA, ranged between 1.6 and 4.1 % dw (Oviatt and Nixon, 1975). The carbon contents in sediment samples (0 - 2 cm) from 13 to 24 m depth in the NW Adriatic coastal area ranged between 0.3 and 0.9 % (Giordani and Angiolini, 1983). Similarly, in sediments from Alaska, collected at 60 m depth, the carbon content amounted to 0.6 % (Henrichs and Doyle, 1986).

2.3.1 Decomposition of sedimentary organic material

Long-term studies of the bacterial decomposition of marine plankton showed that the organic matter decomposed via first-order kinetics could be divided into two decomposable fractions of considerably different reactivity, and a non-metabolizable fraction (Westrich and Berner, 1984). For oxic decomposition of algae, values from 7 to 24 y^{-1} have been reported for the most labile fraction, and 1.4 to 3.2 y^{-1} for a less rapidly metabolized fraction (Table 3).

Table 3

Experimentally determined first-order decay constants for oxic organic matter decomposition at 20-22°C. From Westrich and Berner (1984).

Source	k_1 (yr^{-1})	k_2 (yr^{-1})
Grill and Richards 1964—plankton decomposition under marine conditions; decay constants determined for the decomposition of organic nitrogen and phosphorus.	14	3.2
Jewell and McCarty 1968—algae decomposition under freshwater conditions; decay constants determined for overall organic carbon decomposition.	7–22	
Otsuki and Hanya 1972—algae decomposition under freshwater conditions; decay constants determined for the decomposition of organic nitrogen.	11–26	1.8
Fallon and Brock 1979—algae decomposition under freshwater conditions; decay constants determined for overall organic carbon decomposition.	15	
Ogura 1975—dissolved organic matter in seawater; decay constants for DOC.	3–33	0.3–3
This study.	24	1.4

Similarly, Garber (1984) noted that in most studies of coastal plankton decomposition under oxic conditions, there was a considerable decrease in decomposition rate after 1 - 2 weeks, or after more than 50 % of the organic material had been remineralized. His rate constants were 7 - 70 y^{-1} for the initial, rapid phase and 1 - 7 y^{-1} for the later phase. An essentially non-reactive fraction was also present which made up about 35 % of the original POC; this fraction seemed to be decomposable only on a much longer time scale. For organic matter decomposing in anoxic sediments, the decomposition rates were 0.4 - 1 y^{-1} (Westrich and Berner, 1984).

The derivation of a single first-order decay rate for sedimentary TOC, is consequently, simplistic because TOC is composed of a variety of organic compounds that decompose at different rates. However, in spite of these complexities, first-order decay rates for TOC have been determined in certain environments (e.g. Berner, 1979; Müller and Mangini, 1981) and then provide a useful estimate of what TOC concentrations can be expected in different sedimentary environments. In 8 sediment cores from Lake Superior, decay rates ranged from $3.7 \cdot 10^{-3}$ to $12.8 \cdot 10^{-3}$ y^{-1}, corresponding to half-lives of 54 to 180 years (Johnson and Evans, 1982). The decay rate in sediment less than 100 years old averaged $(8.6 \pm 3.3) \cdot 10^{-3}$ y^{-1}, corresponding to a half-life of 95 ± 45 years. In 51 impoundments in the USA, mostly consisting of man-made lakes, the average TOC decay constant was $2.4 \cdot 10^{-3}$ y^{-1} (Avnimelech et al., 1984). The deviation from the average value was rather small, considering the heterogeneity of the sediments, the errors in sampling, analysis and estimation of deposition rates.

2.3.2 TOC deposition rates

Suess (1980) compiled sediment trap data from throughout the world's oceans and found the vertical flux of TOC to be related to the surface water productivity and water depth ($\geq$ 50 m):

$$C_f = \frac{C_p}{0.0238 \cdot Z + 0.212}$$

where

C_f = flux of TOC (g C m^{-2} y^{-1})
C_p = primary production (g C m^{-2} y^{-1})
Z = water depth (m)

Johnson et al. (1982) found that the equation adequately predicted TOC deposition rates in large lakes. The TOC influx to the lake floor in Lake Superior ranged from 6 to 31 g C m^2 y^{-1} and averaged 13, which is very close to the vertical flux of TOC that would be predicted from sediment trap studies in the oceans.

2.3.3 TOC release from sediments

Avnimelech et al. (1984) calculated the upward flux of carbon from lake sediments. Assuming that a 100 cm deep sediment layer is contributing carbon to the overlying water, the upward carbon flux from a typical sediment (assuming a deposition rate of 3.8 cm y^{-1}) should be 87 g C m^{-2} y^{-1}. Martens and Klump (1981) obtained a combined flux of CH_4+CO_2 of 410 g C $m^{-2}y^{-1}$ for Cape Lookout Bight, USA.

2.3.4 Methane distribution in the water column

The methane distributions from marine and freshwater environments show a reasonable internal consistency (Reeburgh and Heggie, 1977). Marine water column methane concentrations have been summarized by Lamontagne et al. (1973). Methane increases with depth to concentrations of about 7 µM in anaerobic basins such as the Cariaco Trench and the Black Sea. Methane concentrations of 80 µM are reported for Lake Nitinat (Atkinson and Richards, 1967).

Water column methane profiles in lakes are more variable than the marine distributions. Much higher methane concentrations (0.5 - 1.2 mM) than encountered in marine environments have been reported by Weimer and Lee (1973) and Cappenberg (1972). These distributions also show linear methane concentration gradients that approach zero in the metalimnion. Although production of methane in the water column should be possible, the linear methane gradients reported in the water columns suggest minimal methane production there. The water column methane must primarily originate by diffusion from the sediments.

2.3.5 Microbial activity and gas evolution in sediments

As long as free oxygen is available, the net results of respiration and decay are essentially the reverse of photosynthesis. Carbon is released as CO_2, organically combined nitrogen as NO_3^-, and organically combined phosphorus as HPO_4^{2-}. Bottom sediments usually have an "oxygen debt" (Rich, 1975) that is indicated by low or negative redox potentials. The low redox conditions lead to the generation of anaerobic fermentative products, including gases such as CO_2, CH_4, H_2 and H_2S. Anaerobic decay reactions of organic matter are all mediated by bacteria which use the reactions as a source of energy. In general, each reaction is mediated by a specific type of bacteria, and the reactions occur in strict succession, the most important being:

- Denitrification. In denitrification bacteria use the oxygen in nitrate to oxidize organic carbon to CO_2. Hereby they convert nitrate, which is an essential nutrient, into biologically inert molecular N.

- Deamination of amino acids. The produced NH_3 reacts to form NH_4^+, causing a net rise in pH.

- Sulfate reduction. Sulfate reduction is a bacterial reaction in which bacteria use the oxygen in SO_4^{2-} to oxidize organic matter to CO_2, producing sulfide species as a by-product. HS^- will form rather than H_2S if the pH is above 7, and if any reactive Fe compounds are present, the sulfide species will react with them to form solid sulfides.

- Fermentation reactions. The simplest and one of the most important fermentation reactions is CH_4 generation

$$C_{org} + 2H_2O \rightleftarrows CO_2 + CH_4$$

Marine sediments have a gradient-free zone of low methane concentration ($\geqq 0.05$ mM) located between the sediment-water interface and 0.2 - 1 m depth (Emery and Hoggan, 1958; Martens and Berner, 1974). Below this zone, methane accumulates to much higher concentrations (5 - 15 mM) and sometimes reaches partial pressures high enough to permit its escape as bubbles. The sediment profiles from freshwater environments are much less consistent than the marine profiles and vary over a concentration range from << 1 to > 10 mM. The gradient-free low-methane zone has not been observed in freshwater environments (Figure 3).

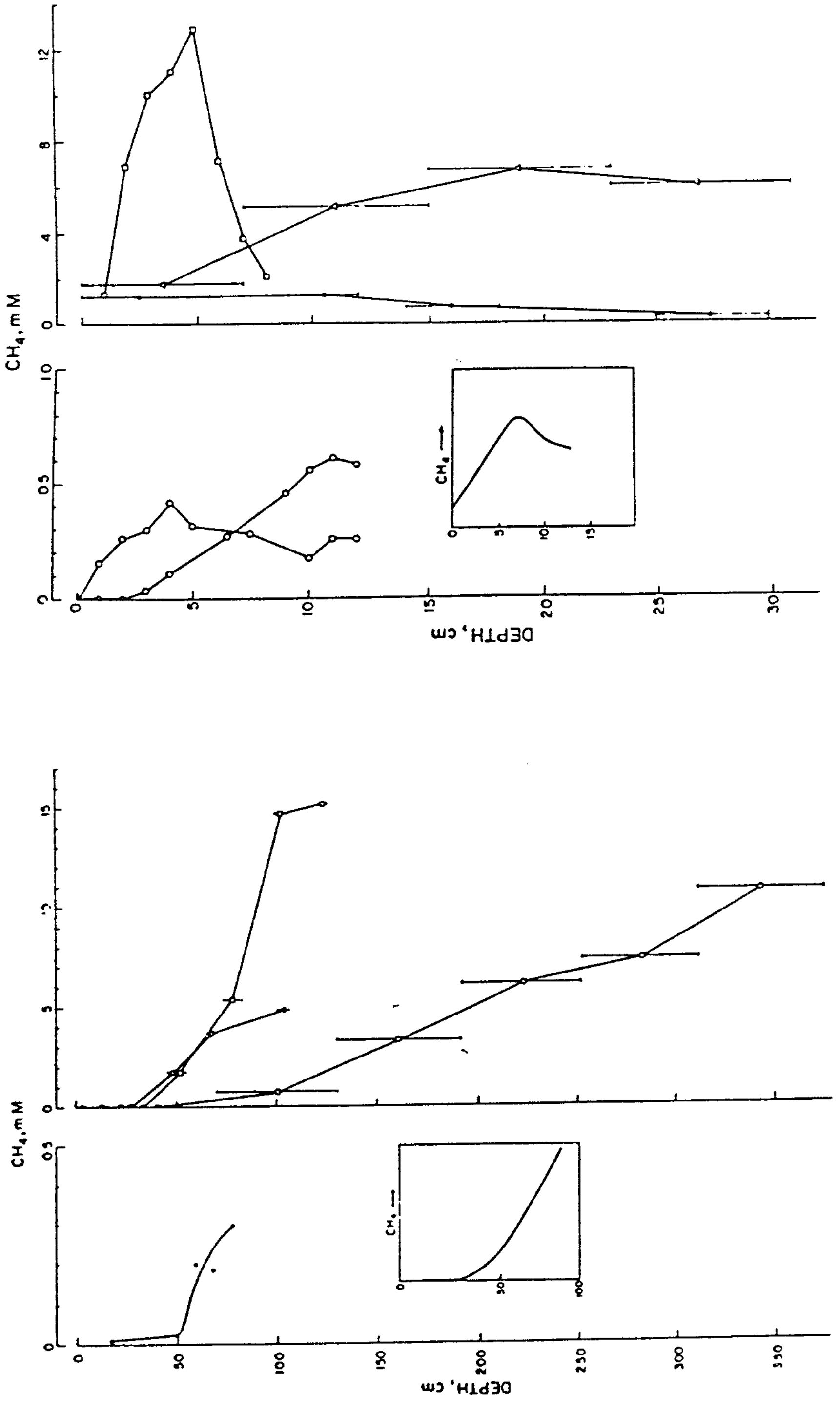

Figure 3

Selected methane profiles in marine and freshwater sediments. From Reeburgh and Heggie (1977).

The distinct differences in the methane distributions from freshwater and marine environments are thus evident, suggesting that some general process must be responsible. Sulfate-reducing and methane-producing bacteria are well known as obligate anaerobes. Cappenberg (1974a, b) demonstrated a commensal relationship between the two bacterial groups. Sulfate reducers require lactate and produce sulfide and acetate, which is a precursor for methane production. Koyama (1964) presented data showing an increase in acetate before methane production began. The onset of methane production has also been linked with the disappearance of sulfate (Claypool and Kaplan, 1974; Martens and Berner, 1974, 1977). Sulfate is never limiting in the water column in marine systems but may become limiting in sediments when consumption by sulfate reducers exceeds the rate of supply from overlying waters (Goldhaber and Kaplan, 1974). Anaerobic methane oxidation in the sediments by sulfate-reducing bacteria produces the low methane surface zone and serves as an effective barrier to transfer of methane across the sediment-water interface. Methane concentrations remain below about 0.1 mM until about 90 % of seawater sulfate is removed (Martens and Berner, 1977). The content of sulphate in brackish water with a salinity of 10 ^{o}/oo gives an oxygen supply about 50 times greater than the supply of dissolved molecular oxygen in water in equilibrium with the atmosphere (Grasshoff and Voipio, 1981). It is therefore easy to understand that the methane formation frequently occurring in lake sediments with a high content of biogenic organic matter has not been observed in, e.g., the Baltic Sea sediments. Summarizing, methane distributions in marine sediments show a low concentration surface zone where concentrations increase with depth. In freshwater environments, the formation of hydrogen sulfide is rare. Although nitrate and sulfate are thermodynamically preferred to organic compounds and carbon dioxide as electron acceptors, the low concentrations of nitrate and sulfate typical of freshwater environments limit their availability for these reactions. Decomposition must then proceed via a scheme whereby substrates are first oxidized to molecular hydrogen and acetate by non-methanogenic bacteria. Thereafter methanogenic bacteria oxidize molecular hydrogen and split acetate to produce methane and carbon dioxide as final products of the overall fermentation.

Methane produced in sediment either remains trapped in the sediment or is released into the water column as dissolved methane or as bubbles. Dissolved methane accumulates in the hypolimnion or is oxidized by methane-oxidizing bacteria. Only 1 - 10 % of the upward methane flux from the sediments in the Cariaco Trench escaped oxidation in the sediments and entered the water column (Reeburgh, 1976). Sulfide concentrations in the surface sediments, where sulfate reduction rates were highest, and in the anoxic water column were high enough to inhibit methane production in either of these locations. The effectiveness of the anaerobic oxidation in the sediments and the absence of methane sources other than the deep parts of the sediments account for the low methane concentrations observed in marine water columns. The concave depth distribution of methane can be explained by methane consumption as it diffuses upward through the zone of sulfate reduction.

Recent investigations have attempted to quantify methane cycling in eutrophic lakes (review by Rudd and Taylor, 1980). Although the methane in sediments can be very intense and reach as much as 85 % of the total gas volume formed there, little of the methane escapes to the atmosphere in low to moderately productive lakes, owing to the activity of methane-oxidizing bacteria in the overlying water column. High rates of methane oxidation, amounting to as much as 95 % of the annual quantity of methane produced, have been observed during lake circulation (Rudd and Hamilton, 1975). A methane budget for the eutrophic Lake 227, western Ontario, was prepared by Rudd and Hamilton (1978). The primary production was 138 g C $m^{-2}y^{-1}$ and the total carbon input to the sediments was 33 g C $m^{-2}y^{-1}$. Of this input, 55 % or 18 g C $m^{-2}y^{-1}$ was regenerated as methane. However, 36 % of the total carbon input or 12 g CH_4C^- $m^{-2}y^{-1}$ was recycled by methane oxidizers to bacterial cell carbon and CO_2. The remaining amount, about 6 g C $m^{-2}y^{-1}$ or 40 % of the annual production of methane was released to the atmosphere. Similarly, Robertson (1979) found that carbon regeneration as methane from the anoxic sediments amounted to 36 % of the carbon deposited onto the sediments of a less productive and 59 % of that deposited in a more productive lake. The relative amounts of POC converted to CH_4^-C in sediments, and losses of CH_4^-C by evasion to the atmosphere are viewed in Table 4 for a number of eutrophic lakes. An amount of organic carbon regenerated as methane equivalent to as much as 60 % of the total carbon input has been found in several very productive lakes. Methane-oxidizing bacteria can recycle about two-thirds of this carbon.

Table 4

Relative amounts of POC converted to methane carbon in profundal sediments, and losses of methane carbon by evasion to the atmosphere in a number of eutrophic lakes. From Wetzel et al. (1983).

LAKE	PERIOD	% OF INPUT C CONVERTED TO CH$_4$	% OF INPUT C LOST FROM LAKE AS CH$_4$−C
Lake 227, Ontario (Rudd and Hamilton, 1978)	Annual	55	<10
Wintergreen Lake, Michigan (Strayer and Tiedje, 1978; Molongoski and Klug, 1980a)	Summer, 1976	34	14
	Summer, 1977	44	20
Third Sister Lake, Michigan (Robertson, 1979)	Summer	36	6
Frains Lake, Michigan (Robertson, 1979)	Summer	59	28
Lake Mendota, Wisconsin (Fallon, et al., 1980)	Summer	54	ca. 5

Values of particulate loading are based on phytoplanktonic sestonic sedimentation only; inputs of organic carbon from the wetlands and littoral productivity, which are extensive in many of these lakes and could be transported along the sediments to hypolimnetic areas and missed in pelagial sedimentation traps, were not evaluated. Therefore, the methanogenetic values could be overestimates.

Bubble formation occurs when saturation concentration is exceeded, and bubble releases to the water surface occur in shallow areas and marshes where intense anaerobic fermentation takes place in the superficial sediments. Released bubbles may escape to the atmosphere and leave the carbon cycle of the lake, or go into solution. Usually, these gases are redissolved and metabolized by bacteria. However, as lakes become very eutrophic, the anaerobic fermentation of organic matter in the sediment can reach levels sufficient to result in the steady ebullition of gases which may constitute a significant portion of the total methane release from the sediment. In the hypereutrophic Wintergreen Lake, Michigan, 21 moles $m^{-2}d^{-1}$ (0.3 g $m^{-2}d^{-1}$) of CH_4, on average, left the anaerobic sediments by ebullition during May to August (Strayer and Tiedje, 1978). Kelly and Chynoweth (1979) showed that the fraction of the total release of methane which escaped via ebullition was 17 and 48 %, respectively, for two eutrophic lakes.

Besides, they reported that about 90 % of the methane released from the sediment as bubbles escaped to the atmosphere. In vitro tests showed that rates of methane production in surface sediments during the summer were directly proportional to the sedimentation rates of organic carbon. The sedimentation rates of organic carbon in the two eutrophic lakes were 390 and 180 mg C $m^{-2}d^{-1}$, respectively. The corresponding methane production rates were 100 and 52 mg C $m^{-2}d^{-1}$, yielding a methane production/sedimentation ratio of 26 and 29 %, respectively. The relation between in-situ measurements of released methane and deposition of organic carbon is shown in Figure 4.

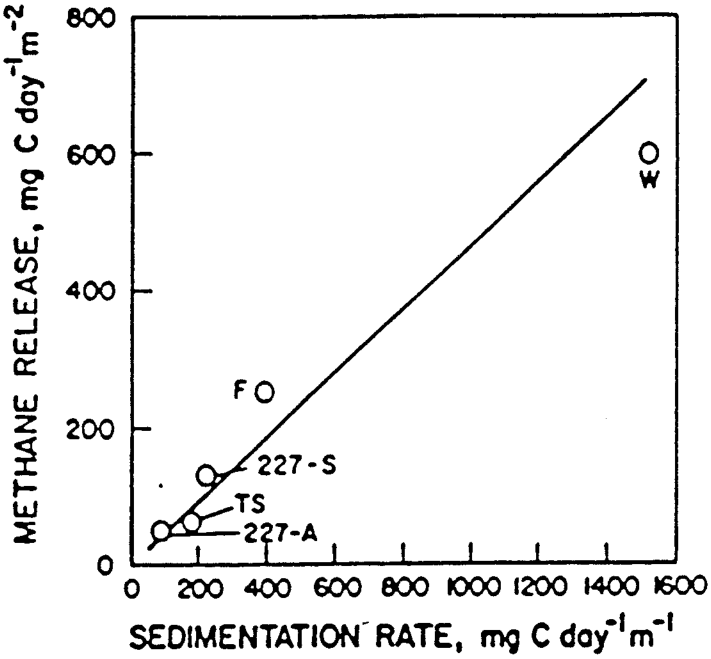

Figure 4

In-situ measurements of methane release rates versus sedimentation rates. From Kelly and Chynoweth (1979).

Gas evolution from marine and freshwater sediments have been reported by a few authors. In the marine environment, Crill and Martens (1981) measured methane evolution at Cape Lookout, N.C, USA, and obtained an average flux of 50 g C $m^{-2}y^{-1}$. In fresh waters, Kelly and Chynoweth (1979) reported a total methane flux of 65 and 227 mg C $m^{-2}d^{-1}$ (24 and 83 g C $m^{-2}y^{-1}$) for two eutrophic lakes. Ebullition contributed 17 and 48 % of the total release, or 11 and 110 g C $m^{-2}d^{-1}$ (4 and 40 g C $m^{-2}y^{-1}$), respectively. The differences in the proportions of methane release, oxidation and ebullition are probably due to physical characteristics of the sediment. In Lake 227, no methane bubbles were recorded, even though methanogenesis occurred in the sediments (Rudd and Hamilton, 1978).

3 Carbon flux in aquatic ecosystems

Quantitative carbon fluxes within marine and freshwater ecosystems have been assessed by several authors. The underlying premise is that the synthesis of organic material begins with the fixation (primarily photosynthetic) of CO_2. Much of this organic carbon is variously cycled within the ecosystem before being respired (primarily via decomposition) and returned as CO_2.

Carbon budgets for three small US and Canadian lakes (Lawrence Lake, Mirror Lake and Lakes 224 and 302) are reviewed. Transfer coefficients between atmosphere, water sediment, fish and terrestrial surroundings are - if sufficient data are available - derived from these budget calculations. Besides, the carbon budged for Lake Ontario, representing a large freshwater basin is presented. Carbon flow and transfer coefficients for Hudson Bay are also given, representing a marine recipient.

3.1 Lawrence Lake

A quantitative representation of the organic carbon exchanges within Lawrence Lake, a small hardwater Michigan lake, was performed by Wetzel et al. (1972) (Figure 5).

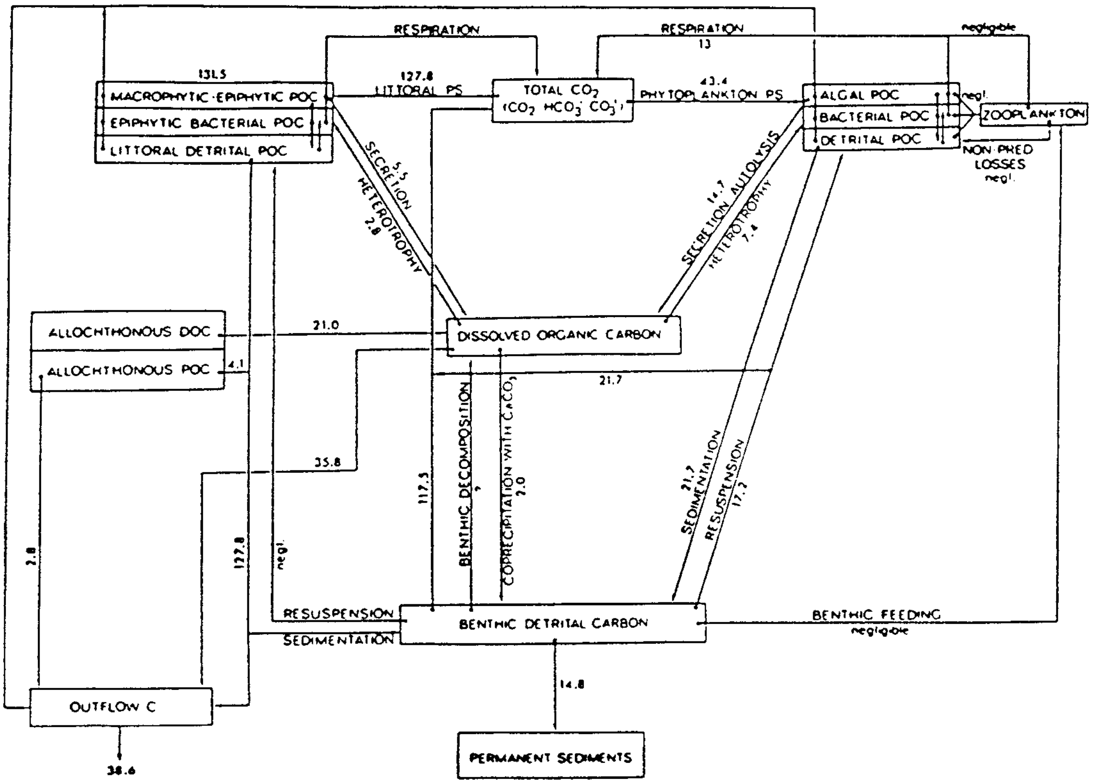

Figure 5

Flux of organic carbon (g m^{-2}y^{-1}) in Lawrence Lake, Michigan, USA. From Wetzel et al. (1972).

Influxes of allochthonous DOC and POC were also analysed in relation to their fate within and losses from the lake (Wetzel and Otsuki, 1974). The in- and outflows of allochthonous DOC were highly significant for the total carbon metabolism of Lawrence Lake, whereas allochthonous influxes and outflows of POC were relatively minor in comparison. The mean value for DOC for all depths and periods was 5.6 ppm with an observed range of 1.5 to 9.6 ppm. The mean POC concentration was 0.47 ppm with a range of 0.05 to 1.7 ppm. An assumed alkalinity of 200 ppm as $CaCO_3$ will give a DIC content of about 48 ppm.

The DOC:POC ratio of ~ 10:1 was very constant over the entire period of study. Groundwater influx contributed almost 40 % of the total yearly input of water, and the annual residence time of the lake water was 0.64 y. The DOC content of the groundwater was very low (~ 1 ppm) but because of the large volume of this inflow, it contributed 30 % to the annual influx of allochthonous DOC (Table 5).

Table 5

Net ecosystem production of organic carbon in Lawrence Lake, michigan, USA. From Wetzel et al. (1972).

	DISSOLVED ORGANIC CARBON	PARTICULATE ORGANIC CARBON	TOTAL (g C m^{-2} YEAR^{-1})
Inputs			
Inlet 1	7.0	2.0	9.0
Inlet 2	7.9	1.0	8.9
Groundwater	6.0	1.2	7.2
Total	20.9	4.2	25.1
Outputs			
Outflow	35.8	2.8	38.6
Sedimentation	-	14.8	14.8
Total	35.8	17.6	53.4

Net ecosystem production (NEP): + 28.3 g C m^{-2} year^{-1}

Groundwater POC was extremely low (~ 0.2 ppm) but on an annual basis constituted over 25 % of the allochthonous POC input. However, allochthonous POC was small in relation to the quantities of littoral and pelagial particulate carbon synthesized within the lake, and represented less than 5 % of the gross production of POC by the lake. Of the autochthonous POC, the major inputs were from the littoral macrophytes and the associated microflora (Table 6).

Table 6

Annual benthic POC budget of Lawrence Lake, Michigan, USA. From Wetzel et al. (1979).

PARTICULATE CARBON	g C m^{-2} $YEAR^{-1}$
Inputs	
Submersed macrophytes	+ 87.9
Epiphytic algae	+ 37.9
Epipelic algae	+ 0.0
Loess	+ 0.0
Precipitation of DOC with $CaCO_3$	+ 2.0
Sedimentation	+ 21.7
Outputs	
Benthic respiration	−117.5
Permanent sedimentation	− 14.8
Balance (utilization)	+ 17.2

POC was either utilized in detrital metabolism or was lost in sedimentation. POC leaving the lake was mainly of autochthonous origin and made up about half the POC entering the lake basin. In contrast, allochthonous DOC represented a major influx of organic carbon. An allochthonous input (DOC + POC) of 25.1 g C m^2y^{-1} (Table 7) multiplied by the lake area $4.96 \cdot 10^4$ m^2 will produce a total allochthonous input of $1.24 \cdot 10^6$ g C y^{-1}.

Table 7

Pelagic DOC budget for Lawrence Lake, Michigan, USA. From Wetzel et al. (1972).

DISSOLVED ORGANIC CARBON	g C m^{-2} YEAR^{-1}	PARTICULATE ORGANIC CARBON	g C m^{-2} YEAR^{-1}
Inputs		**Inputs**	
Algal secretion Algal autolysis	14.7	Phytoplankton Resuspension	43.4 17.2
Littoral secretion:		Allochthonous	4.1
Macrophytes	3.5	Bacteria:	
Epiphytic algae	1.9	Chemosynthesis	7.1
Epipelic algae	0.1	Heterotrophy of	
Allochthonous DOC	21.0	DOC	7.4
Total	41.2	Total	79.2
Losses		**Losses**	
Outflow	35.8	Outflow POC	2.8
Coprecipitation with $CaCO_3$	2.0	Sedimentation	21.7
Total	37.8	Total	24.5
Gross production	+3.4	**Gross production**	+54.7
Respiration		**Respiration**	
Bacteria	−20.6	Algae	13.0
(Estimated as 50% of DOC		Sedimenting POC	21.7
production)		Total	−34.7
Net production	−17.2	**Net production**	+20.0

Total net production (DOC + POC) = +2.8 g C m^{-2} year^{-1}

The drainage area comprised 7.1 times the lake area, and the organic carbon flow from the terrestrial surroundings was then 3.5 g C $m^{-2}y^{-1}$, based on land area. An assumed DIC content of 48 ppm and a yearly outflow of $4.4 \cdot 10^8$ l from Lawrence Lake (Wetzel and Otsuki, 1974), yields 59 g m^{-2} y^{-1}. Thus, totally about 63 g C m^{-2} y^{-1} (based on land area) will be lost from the lake system due to fluvial outputs. Most of the dissolved organic matter was highly refractory to bacterial degradation and was composed of compounds of humic structure that persisted in the lake with relatively long residence times. Considerably more DOC, about twice the amount, left the lake system than entered from allochthonous sources. A net loss of 15 g DOC $m^{-2}y^{-1}$ from the outflow of the lake was observed, which represented a major pathway of removal of organic carbon synthesized within the lake.

The single largest flow of carbon within Lawrence Lake originated as primarily particulate production in the littoral (Table 6). This material was distributed to the deeper benthos where it was supplemented by additional particulate material sedimenting from the pelagic zone (Table 7). Sedimentation of POC to the bottom was 50 % of the total production. Subsequent benthic respiration released a major amount of the imported organic carbon to the pool of dissolved CO_2, representing the primary return flow of carbon to the photic zone. Gross benthic input for one year was 240.8 g C $m^{-2}y^{-1}$. Net input after respiration (117.5 g C $m^{-2}y^{-1}$) was 123.3 g C $m^{-2}y^{-1}$. Only a small fraction, 14.8 g C $m^{-2}y^{-2}$, was permanently lost to the sediment (Rich and Wetzel, 1972) while the remaining amount (123.3 - 14.8 = 108.5 g - C $m^{-2}y^{-1}$) was resuspended, suggesting a major recycling process between the sediments and the water column.

3.1.1 Derivation of transfer coefficients

Lake water - lake surface sediment

The mean organic (DOC+POC) content in lake water was 6.1 ppm. A gross input of 240.8 g C $m^{-2}y^{-1}$, multiplied by the area of the lake, $4.96 \cdot 10^4$ m^2, will give a total benthic input of $1.19 \cdot 10^7$ g C y^{-1}. The lake volume of $2.92 \cdot 10^8$ l gives a total carbon content of $1.58 \cdot 10^7$ g C in the water compartment. The transfer coefficient water-surface sediment is then

$$k = \frac{1.19 \cdot 10^{7}}{1.58 \cdot 10^{7}}$$

$$k = 0.75\ y^{-1}$$

Lake water - outlet

The total fluvial output of organic carbon from Lawrence Lake is 38.6 g C $m^{-2}y^{-1}$ (Table 5).

Adding the DIC content of the water, a total outflow of $2.29 \cdot 10^7$ g C y^{-1} will occur. The inventory of carbon in lake water is $1.58 \cdot 10^7$ g and the transfer coefficient is

$$k = \frac{2.29 \cdot 10^7}{1.58 \cdot 10^7}$$

$$k = 1.45 \ y^{-1}$$

3.2 Mirror Lake

Jordan and Likens (1975) presented an organic carbon balance for Mirror Lake, which is a small, oligotrophic softwater basin in New Hampshire, USA. Mean annual standing stocks in Mirror Lake are presented in Table 8. In many fresh waters the concentration of DIC is greater than DOC. This is not the case for the soft water of Mirror Lake. DIC concentrations ranged from 0.84 to 5.88 ppm with a mean value of 1.53 ppm. DOC ranged from 1.1 to 3.6 ppm (mean value 2.42 ppm) while POC contributed 6.4 times less or 0.38 ppm. Autochthonous primary production was the major source of organic carbon in the lake, accounting for 76 % of the total input (Table 9).

Table 8

Mean annual standing stocks in Mirror lake, Hampshire, USA, expressed in mg C m^{-2}. From Jordan and Likens (1975).

Seston	
Phytoplankton	375[a]
Zooplankton	200[a]
Bacteria	40[c]
Detritus (particulate)	1,500[b]
Bacteria (benthic)	3,000[c]
Benthic invertebrates	700[b]
Macrophytes	1,700[b]
Fish	400[c]
Epilithiphyton	30[c]
Salamanders	5[b]
DIC	8,600[a]
DOC	13,600[a]
Benthic detritus (10 cm)	1,000,000[a]

[a] ± 20 %

[b] ± 50 %

[c] right order of magnitude

Table 9

Annual benthic carbon fluxes in Mirror Lake, New Hampshire, USA. From Wetzel (1983).

COMPONENTS	$g\ C\ m^{-2}$ YEAR^{-1}	PER CENT OF SUBTOTAL
Inputs		
Autochthonous		
Phytoplankton	56.5	69.5
Epilithic algae	2.2	2.7
Epipelic algae	0.6	0.7
Epiphytic algae	0.06	0.07
Macrophytes	2.5	3.1
Dark CO_2 fixation	2.1	2.6
Allochthonous		
With precipitation	1.4	1.7
Shoreline litter	4.3	5.3
Stream DOC	10.5	12.9
Stream POC	1.15	1.4
	81.31	100.0
Outputs		
Respiration		
Phytoplankton	19.1	23.5
Zooplankton	12.0	14.8
Macrophytes	1.0	1.2
Attached algae	1.16	1.4
Benthic invertebrates	2.8	3.4
Fish	0.2	0.2
Sediment bacteria	17.3	21.3
Planktonic bacteria	4.9	6.0
Permanent sedimentation	10.7*	13.2
Outflow		
Dissolved	10.87	13.4
Particulate	0.78	1.0
Insect emergence	0.5	0.6
	81.31	100.0

From unpublished data of M. Jordan, G. E. Likens, and B. Petersen (1982).
*Estimated by difference.

In contrast to the conditions in Lawrence Lake (Wetzel et al., 1972), phytoplankton accounted for 90 % of the autochthonous production whereas macrophytes were of minor importance. The single most important allochthonous input was fluvial DOC (60 %). A total fluvial input of 1 073 mg C m^{-2} y^{-1} and a lake area of 15 ha will give $1.6 \cdot 10^6$ g C y^{-1} as a contribution of organic carbon from runoff. A drainage area of 85 ha will imply an input of 1.9 g C m^{-2} y^{-1}, based on land area. The average inflow due to runoff is $6.5 \cdot 10^6$ l ha^{-1} which gives a total runoff of $5.5 \cdot 10^8$ l y^{-1}. Multiplication by the average DIC content 1.53 ppm will yield about 1 g C m^{-2} y^{-1} from the DIC pool. Accordingly, a total allochthonous inflow of 2.9 g C m^{-2} y^{-1}, based on land area, will occur to Mirror Lake. Of the total organic carbon inputs, 72 % was lost through respiration to the DIC pool, 13 % by permanent sedimentation and 15 % by fluvial outputs.

3.2.1 Derivation of transfer coefficients

Lake water - lake surface sediment

The total carbon content in the lake water was 4.3 ppm. The calculated water volume $8.62 \cdot 10^8$ l gives an inventory of $3.73 \cdot 10^6$ g C in the water mass. The gross input of carbon to the sediment (permanent sedimentation + respiration) amounted to 10.7 + 20.2 = 30.9 g C $m^{-2} y^{-1}$. A total area of $1.15 \cdot 10^5$ m^2 gives a total benthic input of $3.55 \cdot 10^6$ g C y^{-1}. The transfer coefficient water-sediment is then

$$k = \frac{3.55 \cdot 10^6}{3.73 \cdot 10^6}$$

$$k = 0.95 \ y^{-1}$$

Sediment - lake water

The inventory of carbon in the lake sediment was 1 000 g C m^{-1} (Table 8). The benthic bacteria and invertebrates contributed 3 and 0.7 g C m^{-2}, respectively. Benthic respiration amounted to 20.2 g C m^{-2} y^{-1} (Table 9) and the fractional transfer of carbon from the sediment to the water mass is

$$k = \frac{20.1}{1003.7}$$

$$k = 2 \cdot 10^{-2} \ y^{-1}$$

Lake water - outlet

The inventory of carbon in the water mass was $3.73 \cdot 10^6$ g C. A total carbon concentration in the outflow of 18.0 g C m^{-2} y^{-1} (DOC+POC=11.7, DIC=6.3) and a lake area of $1.50 \cdot 10^5$ m^2 will give a transfer coefficient of

$$k = \frac{2.70 \cdot 10^6}{3.73 \cdot 10^6}$$

$$k = 0.72 \ y^{-1}$$

Fish - lake water

The inventory of carbon in the fish pool was 400 mg C m^{-2} (Table 8). Respiration amounted to 0.2 g C m^{-2} y^{-1}, and the transfer coefficient fish-lake water is

$$k = \frac{0.2}{0.4}$$

$$k = 0.5\ y^{-1}$$

3.3 Lake 224

The work by Hesslein et al. (1980) shows in an illustrative way the fate of radiocarbon implanted in a lake ecosystem. C-14 was added as $NaHCO_3$ to the epilimnion of the dimictic oligotrophic Lake no. 224 in the Experimental Lakes Area, Canada. The dominant pathways of C-14 in the epilimnion are shown in Figure 6.

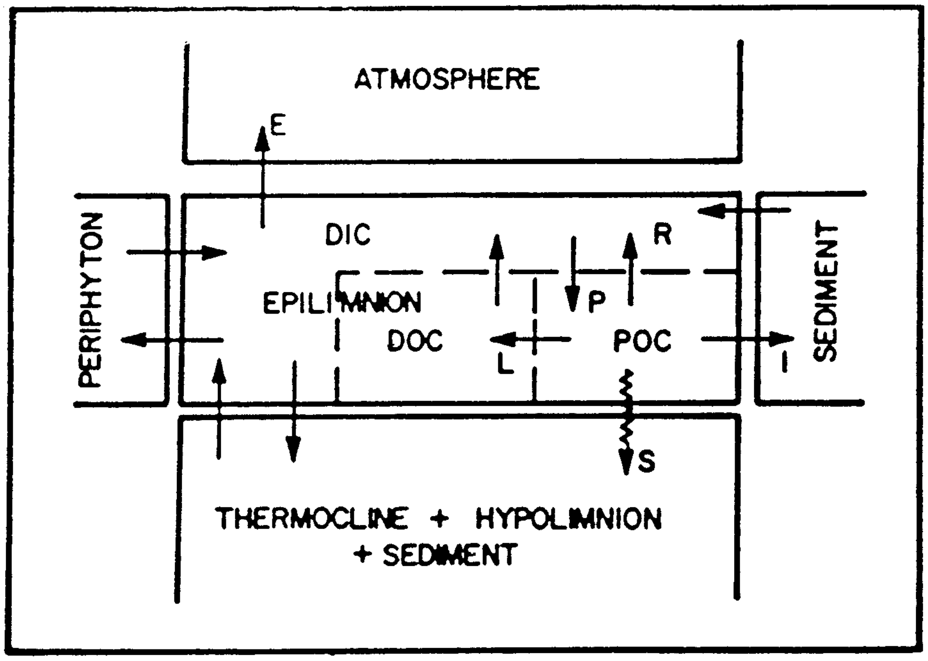

Figure 6

^{14}C pathways in the epilimnion of lake 224, Canada. From Hesslein et al. (1980).

DIC, DOC and POC contributed 1.18, 3.42 and 0.52 ppm, respectively to the content of carbon in the water. These values agreed well with the corresponding concentrations 1.40, 5.65 and 0.92 ppm recorded for Lake 302 (Bower and McCorkle, 1980). The C-14 content of the DIC pool decreased rapidly after addition. The rate of decrease was about 0.053 d^{-1}, or a half-time of about 13 d. C-14 in DOC increased rapidly over the first days, reaching a plateau after 2 weeks. The dissolved CO_2 content was 1.7 times that for equilibrium with the atmosphere. This implies that CO_2 must be leaving the lake at 1.7 times the rate that it is entering. The yearly loss of carbon by net gas evasion was estimated to ~ 24 g C $m^{-2}y^{-1}$. However, the concentration of DIC did not change significantly, and the amount of DIC lost to the atmosphere must, therefore, have been replenished from some other pool in the lake. Three sources exist: net respiration in the epilimnetic water, entrainment of DIC from supersaturated hyplimnetic water, and respiration in the epilimnetic sediment. No consistent change in POC and DOC occurred during the experiment. To produce the CO_2 necessary to replace that lost by the gas evasion, at least 10 % of the combined pool of POC + DOC would be oxidized. This argues against net respiration in the epilimninon.

DIC concentrations in the hypolimnion were no higher than the values for the epilimnion, so entrainment could not have supplied sufficient additional DIC.

Sediment fluxes of DIC from epilimnetic sediments have been measured at $3.2 \cdot 10^{-1}$ g C $m^{-2}d^{-1}$ in a eutrophic lake (Hesslein, 1976). However, this flux is probably greater than could be expected in this oligotrophic lake. Besides, the area of the epilimnetic sediment was about 30 % of the surface area of Lake 224, and the flux of carbon from the sediment might not compensate for losses to the atmosphere.

The rate of photosynthesis was determined from the appearance of C-14 as POC and DOC pools. Calculations showed that 35 % of POC and only 6 % of DOC were involved in the photosynthetic carbon pool of the lake. This led to the conclusion that 65 % of POC and 94 % of DOC were allochthonous.

To summarize, the dominant loss from the DIC pool was via CO_2 evasion to the overlying atmosphere. The next most important loss from the DIC pool was by photosynthetic fixation of inorganic carbon by epilimnetic phytoplankton. About half of the C-14 thus incorporated into the POC pool was converted into soluble organic molecules which became part of the epilimnetic DOC pool. The amount of C-14 lost to the sediments of the epilimnion, to the hypolimnion and to macrophyte biomass was not significant for the C-14 mass balance. The net loss of carbon to the atmosphere demands that carbon fixed in photosynthesis be derived from a source other than the atmosphere. Two possible sources for this carbon are the sediments and inflow from external sources. If the sediments are net accumulators of carbon, as suggested by Kipphut (1978), the only source is inflow. The carbon added by runoff must be organic, because DIC levels in runoff are very low due to the absence of carbonate minerals in this area. Such an addition of terrestrial organic material could explain the large fraction of POC and DOC which apparently is not a result of photosynthesis in the lake.

3.3.1 Derivation transfer coefficients

Water - atmosphere

The mean CO_2 evasion from the lake surface was 24 $g\ m^{-2}\ y^{-1}$. Multiplication with the lake area $2.59 \cdot 10^5\ m^2$ yields a total annual loss of $6.22 \cdot 10^6$ g C y^{-1}. An estimated water volume of $3 \cdot 10^9$ l and a total carbon content of 5.1 ppm will give an inventory of $1.53 \cdot 10^7$ g C in the water mass. The transfer coefficient water-atmosphere becomes

$$k = \frac{6.22 \cdot 10^6}{1.53 \cdot 10^7}$$

$$k = 0.41\ y^{-1}$$

3.4 Lake 302

The carbon budget for a Canadian oligotrophic lake (L 302) was analysed by Bower and McCorkle (1980) adding C-14 as a tracer to the water inside a large plastic enclosure. A large outflux of CO_2 was recorded and the net mass of carbon lost from the water surface was

$0.41\ g\ C\ m^{-2}\ d^{-1}$ for a 16 d period. Uptake by periphyton was negligible and the C-14 content of the POC from sediment traps was less than 0.4 % of the original amount added to the enclosure, or $0.2\ g\ C\ m^{-2}$ over 16 d. There was therefore no significant loss of C-14 other than gas exchange.

3.5 Lake Ontario

Robertson and Eadie (1975) estimated the carbon budget for Lake Ontario. Groundwater and autochthonous sources are quantitatively insufficient (Eadie and Robertson, 1976). The rivers contributed 63 % to the total carbon influx, while primary production made up the rest. The estimates by Robertson and Eadie (1975) also showed that a strong outflux of CO_2 occurred through the lake surface during winter followed by a weak influx during the spring and the summer. The large flux of carbon lost during the winter resulted from water strongly supersaturated with CO_2 being brought to the surface during the mixing of the entire water mass in this season. The supersaturation of CO_2 in the hypolimnetic water was the result of decomposition of sedimentary organic material. The net output of carbon from the lake surface to the atmosphere amounted to about one-third of the total output, while loss of river water constituted 63 % and permanent sedimentation only 3 %. The in- and outflow terms approximately balanced each other.

3.5.1 Derivation of transfer coefficients

Lake water - atmosphere

The gas evasion from the lake surface to the atmosphere was 34 % of the total outflow, implying a loss of $2.8 \cdot 10^{12}\ g\ C\ y^{-1}$. The mean carbon content in Lake Ontario water was 25.2 ppm, and the water volume $1.64 \cdot 10^{12}\ m^3$, which gives an inventory of $4.13 \cdot 10^{13}\ g\ C$. The transfer coefficient lake water-atmosphere is

$$k = \frac{2.80 \cdot 10^{12}}{4.13 \cdot 10^{13}}$$

$$k = 6.8 \cdot 10^{-2}\ y^{-1}$$

Lake water - sediment

Assuming a whole-lake sedimentation rate of $8.36 \cdot 10^{12}$ g y^{-1} and a total carbon content in superficial sediment of 3.6 % (McKee, Rowsell, 1984), the flux of carbon to sediments in Lake Ontario is $3.01 \cdot 10^{11}$ g C y^{-1}. The carbon content in lake water is $4.13 \cdot 10^{13}$ g and the fractional transfer rate from water to sediment is

$$k = \frac{3.01 \cdot 10^{11}}{4.13 \cdot 10^{13}}$$

$$k = 7.3 \cdot 10^{-3} \quad y^{-1}$$

Lake water - outlet

The summed river outflows carried 64 % of the total annual output or $5.30 \cdot 10^{12}$ g C. A total carbon inventory of $4.1 \cdot 10^{13}$ g C in the water mass gives the transfer coefficient lake water-outlet

$$k = \frac{5.30 \; 10^{12}}{4.13 \cdot 10^{13}}$$

$$k = 0.13 \; y^{-1}$$

3.6 Hudson Bay

McKee and Rowsell (1984) calculated carbon inventories and transfer coefficients for the Hudson Bay ecosystem. Due to a large influx of freshwater, Hudson Bay generally contains water of relatively low salinity in the surface layer, and denser water of high salinity in the underlying water mass. In seawater of 35 o/oo salinity, inorganic carbon levels are usually about 28 ppm (Riley and Chester, 1971). Assuming proportionality between salinity and inorganic carbon, the estimated inorganic carbon content in Hudson Bay water is 24 ppm. DOC concentrations in seawater are usually in the range of 0.3 to 3 ppm and are generally 10 to 20 times greater in concentration than POC (Riley and Chester, 1971). Assuming that DOC levels are 15 times greater than POC levels, a mean organic carbon contant of 0.9 ppm was supposed to be valid. Summing the inorganic and organic forms of carbon, a total concentration of 24.9 ppm was calculated for Hudson Bay water.

Hudson Bay has a very low rate of primary production and low concentrations of suspended material. Anderson and Roff (1980) observed mean POM concentrations in Hudson Bay of 0.053 ppm for offshore areas and 0.074 ppm for inshore areas. Thus, there is little sedimentation due to in-situ production. However, the large river influence causes considerable input of sedimenting material. Assuming a bulk density of 1.2, a sedimentation rate of 120 g m^{-2} y^{-1} was adopted for Hudson Bay.

In a global model developed by Nair (1981), an atmosphere to surface carbon flux of about 340 g C m^{-2} y^{-1} was calculated (McKee and Rowsell, 1984). In order to account for ice cover, the flux was reduced by 25 %, meaning a water to atmosphere flux of 258 g C m^{-2} y^{-1}. Nair (1981) also estimated an annual flux from cold surface ocean to atmosphere of about 210 g C m^{-2}. Again, assuming a 25 % reduction in flux rate, a value of 158 g C m^{-2} y^{-1} was adopted for Hudson Bay.

3.6.1 Derivation of transfer coefficients

Water - sediment

An organic content of 2.5 % was adopted for the superficial sediments in Hudson Bay. Because 56 % of the organic material in ocean sediments is carbon (Sverdrup et al., 1942), the organic carbon content in Hudson Bay sediments is 1.4 %. A sedimentation rate of 120 g m^{-2} y^{-1} gives an organic carbon flux of about 2 g C m^{-2} y^{-1} or, for the entire area of $7.4 \cdot 10^{11}$ m^2, a total organic carbon flux of $1.4 \cdot 10^{12}$ g C y^{-1}. In comparison, Nair (1981) adopted a sedimentary carbon flux of about 5 g C m^{-2} y^{-1} for "cold ocean" conditions. The total carbon inventory in Hudson Bay water was $2.2 \cdot 10^5$ g C (McKeen and Rowsell, 1984). The transfer coefficient water-sediment for the Hudson Bay area is then

$$k = \frac{1.4 \cdot 10^{12}}{2.2 \cdot 10^{15}}$$

$$k = 6.4 \cdot 10^{-4} \quad y^{-1}$$

Sediment - water

The sedimentation compartment was assumed to be 50 cm deep. Carbon in deeper sediments was treated as lost in the circulation. Assuming a linear decline in organic carbon with depth to a level of 50 % of surface concentrations at 50 cm, the average organic content in the sediment compartment is equivalent to 75 % of surface concentrations. The organic carbon content of the Hudson Bay sediment compartment is then 1.1 %.

The average inorganic carbon level in terrigenous sediment is about 0.21 %. Riley and Chester (1971) indicated no return of inorganic carbon from sediment to water. Thus, the inorganic carbon level was assumed to be constant with depth. The average total carbon content in Hudson Bay sediment was calculated by adding the mean organic and inorganic carbon levels. The inventory of carbon in the Hudson Bay sediment compartment was then 1.3 % ($7.9 \cdot 10^{3}$ g C m^{-2}) or $5.8 \cdot 10^{15}$ g C for the entire bay.

As mentioned above, organic carbon levels in sediments were assumed to decrease by 50 % after burial to a depth of 50 cm. Thus, 50 % of the sedimentary organic carbon was assumed to return to surface water circulation. The rate of organic carbon sedimentation is then

$$1.4 \cdot 10^{12} \cdot \left(\frac{1.4}{1.4+0.2}\right) = 1.2 \cdot 10^{12} \text{ g C y}^{-1}$$

A sedimentation rate of 100 mm/1000 y was adopted for Hudson Bay. Half of these fluxes was returned to surface water after sediment burial to 50 cm depth, or every 5000 y. The sediment to water carbon flux is then

$$\frac{1.2 \cdot 10^{12}}{5000} \cdot 0.50 = 1.2 \cdot 10^{8} \text{ g C y}^{-1}$$

the corresponding transfer coefficient is

$$k = \frac{1.2 \cdot 10^{8}}{5.9 \cdot 10^{15}}$$

$$k = 2.1 \cdot 10^{-8}\ y^{-1}$$

3.7 Carbon fluxes - conclusions

Quantitative estimations of the carbon flux through marine and freshwater environments generally show that:

- In fresh waters, DOC constitutes the primary source of allochthonous organic carbon loading.
- Within the lake ecosystem itself, the littoral zone is often the dominant source of autochthonously synthesized organic carbon.
- POC is ultimately deposited at the bottom in the static water. The decomposition of organic matter by benthic metabolism often results in a hypolimnetic supersaturation of CO_2 and a subsequent net gas evasion during periods of turbulence.
- Anaerobic fermentation processes in fresh waters result in CH_4 production but only a small gas evasion direct to the atmosphere.

Data concerning the calculated transfer coefficients are compiled in Table 10. For comparison, the carbon fluxes and transfer coefficients calculated by McKee and Rowsell (1984) for representative freshwater and marine recipients in Canada are presented (Tables 11, 12, and 13).

Table 10

Average carbon transfer coefficients (y^{-1}) calculated for three small lakes (Lawrence l., Mirror L., L. 224), one large lake (L. Ontario), and a marine bay (Hudson Bay).

	Transfer coefficient (y^{-1})				
Pathway	Lawrence Lake	Mirror Lake	Lake 224	Lake Ontario	Hudson Bay
water - atmosphere			0.20	0.07	
water - sediment	0.75	0.43		0.007	$6\ 10^{-4}$
sediment - water		0.02			$2\ 10^{-8}$
water - outlet	1.45	0.72		0.13	

Table 11

Fluxes and transfer coefficients (y^{-1}) for carbon in a generalized local freshwater ecosystem, Canada. From McKee and Rowsell (1984).

Compartment		Flux F (g/ha.a)	Fractional Transfer Rate k (a^{-1})
From	To		
Atmosphere	Surface water	5×10^5	4.2×10^{-2}
Surface water	Atmosphere	5×10^5	3.3×10^{-1}
Surface water	Sediment	4.4×10^5	2.9×10^{-1}
Sediment	Surface water	3.8×10^5	1.5×10^{-2}
Surface water	Biota (fish)	1.5×10^2	9.9×10^{-5}
Surface water	Regional water	4.8×10^7*	3.2×10^1

* F and k are calculated on a whole-lake basis (i.e. 100 ha) for flux ro regional waters

Table 12

Fluxes and transfer coefficients (y^{-1}) for carbon in the Lake Nipigon - Great lakes area, Canada. From McKee and Rowsell (1984).

From	To	Flux (g/a)	Fractional Transfer Rate (a^{-1})
Atmosphere:	Surface Water		
- Nipigon		6.8×10^{11}	-
- Superior		1.2×10^{13}	-
- Huron		8.9×10^{12}	-
- Erie		3.9×10^{12}	-
- Ontario		2.9×10^{12}	-
Surface Water:	Atmosphere		
- Nipigon		7.2×10^{11}	1.8×10^{-1}
- Superior		1.3×10^{13}	9.3×10^{-2}
- Huron		9.5×10^{12}	1.3×10^{-1}
- Erie		4.1×10^{12}	3.4×10^{-1}
- Ontario		3.0×10^{12}	7.3×10^{-2}
Surface Water:	Downstream		
- Nipigon	- Superior	2.2×10^{11}	5.6×10^{-2}
- Superior	- Huron	7.7×10^{11}	5.5×10^{-3}
- Huron	- Erie	3.5×10^{12}	4.7×10^{-2}
- Erie	- Ontario	4.4×10^{12}	3.7×10^{-1}
- Ontario	- Global Oceans	5.3×10^{12}	1.3×10^{-1}
Surface Water:	Sediment		
- Nipigon		2.4×10^{10}	6.2×10^{-3}
- Superior		4.4×10^{11}	3.2×10^{-3}
- Huron		1.1×10^{12}	1.5×10^{-2}
- Erie		1.3×10^{12}	1.1×10^{-1}
Ontario		3.0×10^{11}	7.3×10^{-3}
Sediment:	Surface Water		
- Nipigon		1.2×10^{8}	4.4×10^{-5}
- Superior		2.2×10^{9}	4.4×10^{-5}
- Huron		1.2×10^{9}	4.4×10^{-5}
- Erie		2.4×10^{9}	2.5×10^{-5}
- Ontario		5.3×10^{8}	1.3×10^{-5}
Surface Water:	Biota (fish)		
- Nipigon		2.4×10^{7}	6.2×10^{-6}
- Superior		2.8×10^{8}	2.0×10^{-6}
- Huron		2.0×10^{8}	2.7×10^{-6}
- Erie		1.7×10^{9}	1.4×10^{-4}
- Ontario		7.1×10^{7}	1.7×10^{-6}

Table 13

Fluxes and transfer coefficients 8y^{-1}) for carbon in the Hudson Bay area. From McKee and Rowsell (1984).

From	To	Flux (g/a)	Fractional Transfer Rate (a^{-1})
Atmosphere:	Surface Water		
- Albany River		7.5×10^{10}	-
- James Bay		1.9×10^{13}	-
- Hudson Bay		1.9×10^{14}	-
Surface Water:	Atmosphere		
- Albany River		4.7×10^{11}	3.4×10^{-2}
- James Bay		1.2×10^{13}	2.7×10^{-1}
- Hudson Bay		1.2×10^{14}	5.5×10^{-2}
Surface Water:	Downstream		
- Albany River	- James Bay	9.4×10^{11}	6.7×10^{-2}
- James Bay	- Hudson Bay	6.6×10^{12}	1.6×10^{-1}
- Hudson Bay	- Global Oceans	1.8×10^{13}	8.0×10^{-3}
Surface Water:	Sediment		
- Albany River		0	0
- James Bay		9.7×10^{11}	2.3×10^{-2}
- Hudson Bay		1.4×10^{12}	6.4×10^{-4}
Sediment	Surface Water		
- Albany River		0	0
- James Bay		4.4×10^{8}	6.0×10^{-7}
- Hudson Bay		1.2×10^{8}	2.1×10^{-8}

4 Model structure with reference to the SFR site

The inland scenario covers four different areal zones (Bergström and Puigdomenech, 1987). The local zone constitutes a well and a small patch of ground (100 m^2) for the cultivation of vegetables. The water from the well is used for drinking and for irrigation of the patch with totally 200 mm y^{-1}. The local zone is only used for the calculation of individual doses originating from the well water. The regional zone consists of 20 km^2 area in which a 2.5 km^2 large lake constitutes the primary recipient for ^{14}C from the repository. Of the land area, 0.1 km^2 is used for agricultural purposes. Crops were assumed to be raised using sprinkler irrigation, with the irrigation water (200 mm y^{-1}) obtained from the contaminated lake. The lake water discharges into the Baltic Sea which makes up the intermediate zone. Finally, the global zone includes compartments of importance for the long-term dispersion of ^{14}C. The conceptualization of the model into compartments is visualized in Figure 7. The carbon masses of the compartmens are compiled in Table 14.

Figure 7

^{14}C model structure

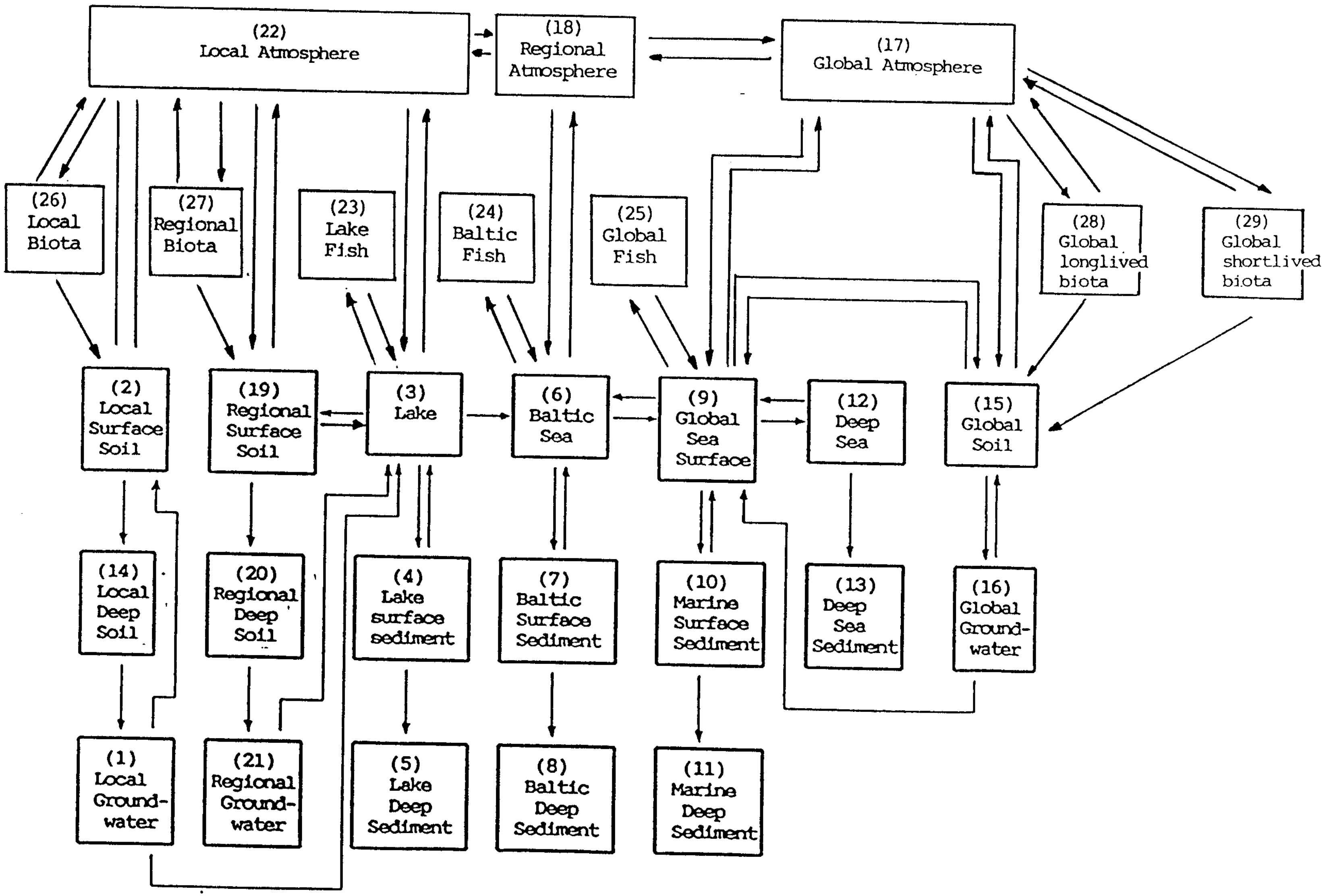

Table 14

Masses of the model compartments

no	compartment	Carbon mass (tons)
1	local groundwater	1*
2	local surface soil	1.15
3	lake water	$8.2 \cdot 10^{1}$
4	lake surface sediment	$3 \cdot 10^{3}$
5	lake deep sediment	1*
6	Baltic Sea water	$4.58 \cdot 10^{8}$
7	Baltic surface sediment	$1.8 \cdot 10^{8}$
8	Baltic deep sediment	1*
9	global sea surface	$1.5 \cdot 10^{12}$
10	marine surface sediment	$3.6 \cdot 10^{9}$
11	marine deep sediment	1*
12	global deep sea	$3.5 \cdot 10^{13}$
13	deep sea sediment	1*
14	local deep soil	1*
15	global soil	$2 \cdot 10^{12}$
16	global groundwater	$6.0 \cdot 10^{18}$
17	global atmosphere	$5.5 \cdot 10^{11}$
18	regional atmosphere	$5.5 \cdot 10^{7}$
19	regional surface soil	$1.1 \cdot 10^{3}$
20	regional deep soil	1*
21	regional groundwater	1*
22	local atmosphere	$4.2 \cdot 10^{2}$
23	lake fish	$1.2 \cdot 10^{-1}$
24	Baltic fish	$2 \cdot 10^{5}$
25	global fish	$1.7 \cdot 10^{9}$
26	local biota	$4 \cdot 10^{-3}$
27	regional biota	5
28	global long-lived biota	$8.3 \cdot 10^{11}$
29	global short-lived biota	$8.3 \cdot 10^{9}$

(*) of no interest in this study

The transport pathways associated with a release of carbon to well and lake water includes drinking water, fish, food crop and animal product ingestion, and inhalation. The exposure pathways for the critical group in the well scenario as well as for the lake scenario are given in Figures 8 and 9, respectively.

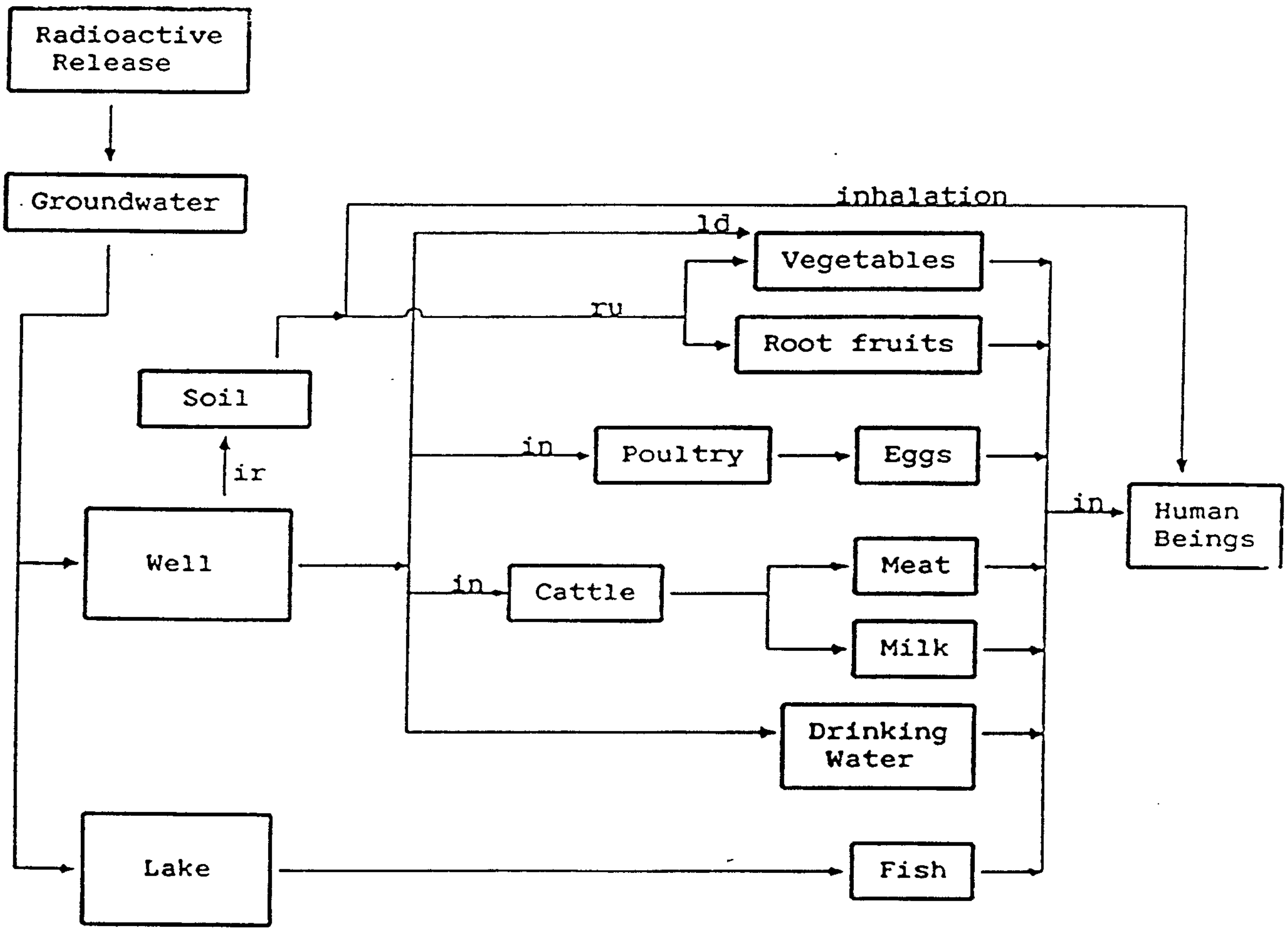

Figure 8

Exposure pathways for the critical group in the well scenario. From Bergström and Puigdomenech (1987).

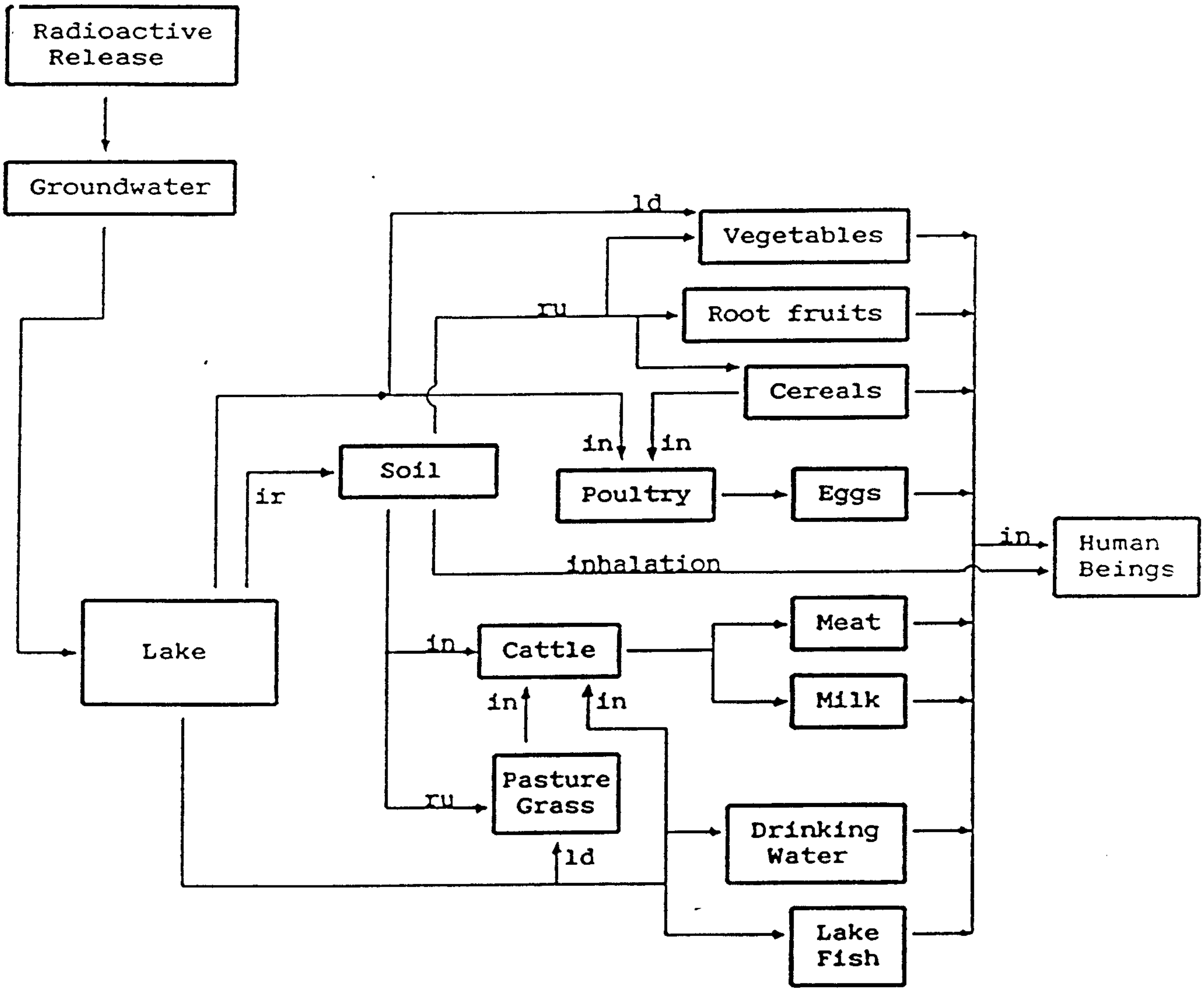

Figure 9

Exposure pathways for the critical group in the lake scenario and for the collective doses in the two land scenarios. From Bergström and Puigdomenech (1987).

The exposure pathways for the regional population are the same as for the critical group in the lake scenario. The exposure route for the intermediate zone is fish consumption. Besides, a possible future harvest of algae is added to the fish yield. For the global area, 10^{10} individuals are assumed to be exposed to all the previously mentioned pathways. Consumption data per capita and estimated population sizes for the different zones are summarized in Tables 15 and 16, respectively.

Table 15

Consumption data. From Bergström and Puigdomenech (1987).

	Zones					
	Local:		Regional:			
	Coast	Inland	Coast	Inland	Intermediate	Global
Individuals						
Inhalation, m^3/year	0.	9438.	0.	9438.	0.	9438.
Drinking water, l/year	0.	600.	0.	600.	0.	600.
Milk, l/year	0.	190.	0.	190.	0.	90.
Meat, kg/year	0.	55.	0.	55.	0.	25.
Eggs, kg/year	0.	198.	0.	198.	0.	220.
Green vegetables, kg/year	0.	25.	0.	25.	0.	90.
Cereals, kg/year	0.	75.	0.	75.	0.	120.
Root-fruits, kg/year	0.	75.	0.	75.	0.	80.
Fish, kg/year	30.	30.	30.	30.	30.	10.
Seaweed, kg/year	0.	0.	0.	0.	10.	10.
Cattle	All zones					
Drinking water, l/day	90.					
Forage, kg/day	16.					
Soil, kg/day	0.3					
Poultry						
Drinking water, l/day	0.25					
Cereals, kg/day	0.11					

Table 16

Estimated population sizes. From Bergström and Puigdomenech (1987).

LOCAL ZONE, inland scenario	
Exposure pathway	Number of individuals
Inhalation	350
Drinking water	350
Milk consumption	160
Meat consumption	20
Vegetables	100
Cereals	370
Root-fruits	140
Egg	200
Fish	60

LOCAL ZONE, coast scenario	
All exposure pathways:	28 000 persons

BALTIC ZONE	
Exposure pathway:	Number of individuals
Fish consumption	$1.2 \cdot 10^7$
Marine plant consumption	$1.2 \cdot 10^6$

GLOBAL ZONE	
Exposure pathway:	Number of individuals:
Marine consumption	10^9
All other pathways	10^{10}

4.1 Inventories of carbon in compartments

Local zone

Local_atmosphere (22)

The local atmosphere was assumed to contain 330 ppm CO_2 or 162 mg C m^{-3} (McKee and Rowsell, 1984). For a surface area of 2.6 km^2 (local surface soil + regional surface soil + local lake), and an air column of 1 000 m height, the carbon inventory is $4.21 \cdot 10^2$ tons.

Local_surface_soil (2)

The local surface soil compartment consists of a 100 m^2 patch used for agricultural purposes (Bergström and Puigdomenech, 1987). The mass is $4.6 \cdot 10^4$ kg. Assuming a mean organic content of 4.3 % dw, and a carbon content of 58 % (McKee and Rowsell, 1984), the inventory of carbon is 1.15 tons.

Local_biota (26)

The biomass of green vegetables in the local biota compartment is supposed to be 1.5 kg m^{-2} ww (Bergman et al., 1979). A carbon content of 50 % dw and a wet/dry weight ratio of 18 will give a carbon content of 42 g m^{-2}. The inventory of carbon is then $4.17 \cdot 10^{-3}$ tons.

Regional zone

Regional_surface_soil (19)

The area of the regional surface soil compartment is 0.1 km^2 and the mass is $4.6 \cdot 10^7$ kg (Bergström and Puigdomenech, 1987). Assuming a mean organic content of the soil of 4.3 % dw, and a carbon concentration of 58 % (McKee and Rowsell, 1984), the inventory of carbon is $1.1 \cdot 10^3$ tons.

Regional_biota (27)

In the regional surface soil compartment, 0.1 km^2 is assumed to be used for agricultural purposes. A mean biomass of 1 320 kg ha^{-1} dw (corn), and a carbon content of 49.6 % dw (McKee and Rowsell, 1984) will give a carbon inventory of $6.55 \cdot 10^6$ g C dw. A ww/dw ratio of 1.3 gives a biomass of 1 716 kg ha^{-1} and an inventory of 5.0 tons of carbon.

Lake (3)

The lake volume is $7.5 \cdot 10^9$ l and the assumed mean total carbon content is 11 ppm, implying a carbon inventory of 82 tons. The surrounding drainage area covers 17.5 km^2. Using the proportions 0.1 km^2 and $1.1 \cdot 10^9$g C, given for the regional surface soil compartment, the carbon content of the drainage area is $1.93 \cdot 10^5$ tons.

Lake_surface_sediment (4)

A bottom area of 2.5 km^2, a sediment depth of 0.1 m, a density of 1.2, a mean carbon content of 5 % dw, and a water content of 80 % will give an inventory of $3 \cdot 10^3$ tons of carbon in the lake surface sediment.

Lake_fish (23)

The biomass of fish in the lake is assumed to be 10 kg ha^{-1} or 2 500 kg for the whole lake. A carbon content of 5 % ww will give an inventory of 0.1 tons.

Regional_atmosphere (18)

The mass of the regional atmosphere compartment is $4.4 \cdot 10^{14}$ kg. Adopting a mean CO_2 concentration of 162 mg m^{-3}, the inventory of carbon becomes $5.5 \cdot 10^7$ tons.

Intermediate zone

Baltic_Sea (6)

A total carbon content in Baltic Sea water of 21 ppm and a volume of 21 800 km^3 yields a carbon inventory of $4.58 \cdot 10^8$ tons.

Baltic_surface_sediment (7)

The following data were supposed to be valid for the Baltic Sea surface sediment: Area $3.7 \cdot 10^5$ km^2, sediment depth 0.1 m, density 1.2, mean carbon content 2 % dw, water content 80 %. This will give a sediment mass of $4.4 \cdot 10^{10}$ tons and a carbon inventory of $1.8 \cdot 10^8$ tons.

Baltic_fish (24)

The total biomass of the commercially exploited fish stocks of the Baltic Sea amounts to about $4.1 \cdot 10^6$ tons, (Thurow, 1980) and the annual yield is about $9 \cdot 10^5$ tons. A mean carbon content of 5 % ww will give an inventory of $2.0 \cdot 10^5$ tons.

Global zone

Global_atmosphere (17)

The global atmosphere is presented by a single well-mixed compartment. A mean carbon concentration of 162 mg m^{-3} (McKee and Rowsell, 1984) and a mass of $4.4 \cdot 10^{18}$ kg (Bergström and Puigdomenech, 1987) will imply an inventory of $5.5 \cdot 10^{11}$ tons of carbon. This figure is somewhat low compared with estimates made by other authors (604 GT by Nair, 1981; 616 GT by Killough, 1980; 631 GT by Hoffert et al., 1981).

Global_soil (15)

Estimates of the global soil pool are uncertain and range $(1 - 3) \cdot 10^{12}$ tons of carbon (Keeling, 1973; Olsen et al., 1978).

Global biota (28, 29)

The biomass of terrestrial primary products is $7 \cdot 10^{11}$ tons of carbon, with an estimated uncertainty of about ± 20 % (Whittaker and Likens, 1975). Net primary production is estimated to about $6.3 \cdot 10^{10}$ tons y^{-1}, 20 % of this being accumulated as wood. Likewise, a total plant mass of carbon of $8.3 \cdot 10^{11}$ tons, and a net production of $5.3 \cdot 10^{10}$ tons was assessed by Woodwell et al. (1978). More than 90 % of the carbon incorporated in terrestrial biota resides in long-lived biota with a mean carbon turnover time of about 100 years (Machta, 1973; Bacastrow and Keeling, 1973) while in short-lived plants, used for human consumption, the mean carbon turnover time is about two years. Less than 1 % of the total plant mass and 8 % of the net production is assigned to cultivated land (Woodwell et al., 1978). A carbon content of $8.3 \cdot 10^{11}$ tons and $8.3 \cdot 10^{9}$ tons was assigned to the global long-lived and short-lived biota, respectively.

Global sea surface (9) and deep sea (12)

The total surface area of the oceans is $3.61 \cdot 10^{14}$ m^2 and the total volume $1.42 \cdot 10^{18}$ m^3 (Dietrich and Kalle, 1965). A mean carbon concentration of 28 ppm will imply an inventory of $3.98 \cdot 10^{13}$ tons, which can be compared with the value $3.65 \cdot 10^{13}$ tons used by Killough (1980), and $3.8 \cdot 10^{13}$ tons by Emanuel et al. (1984). Due to the very different ventilation times of carbon in the well mixed layer and the deep sea region, these two volumes are treated as separate compartments, containing about $1.5 \cdot 10^{12}$ and $3.5 \cdot 10^{13}$ tons of carbon, respectively (Killough, 1980).

Marine surface sediment (10)

The mass of the global marine surface sediment compartment is $2.4 \cdot 10^{12}$ tons (Bergström and Puigdomenech, 1987). An assumed mean carbon content of 0.1 % dw will imply a carbon pool of $3.6 \cdot 10^{9}$ tons.

4.2 Derivation of transfer coefficients

Local atmosphere - local surface soil (22, 2)

Atmosphere to soil flux of carbon occurs via precipitation. Average total carbon content in precipitation is 3.2 ppm (Hutchinson, 1975; Gosz et al., 1978). For an annual rainfall of 600 mm, the carbon flux is 1.9 g C $m^{-2}y^{-1}$. The transfer coefficient atmosphere-soil is then

$$k_{22,\ 2} = \frac{1.9 \cdot 100}{4.21 \cdot 10^{8}}$$

$$k_{22,\ 2} = 4.51 \cdot 10^{-7}\ y^{-1}$$

Local surface soil - local atmosphere (2, 22)

Carbon flux from surface soil to atmosphere occurs due to efflux of CO_2 produced in the soil by bacterial degradation and root respiration. In forest soil, CO_2 respiration varied from 0.6 to 3.6 g C $m^{-2}d^{-1}$ during a one-year investigation (Antos, 1978, quoted in Rau, 1978). A flux rate of $4.2 \cdot 10^{6}$ g C $ha^{-1}y^{-1}$ (1.2 g C $m^{-2}d^{-1}$) was assessed for an agricultural ecosystem within the Canadian shield by McKee and Rowsell (1984). Adopting this flux for Swedish conditions, the transfer coefficient becomes

$$k_{2,\ 22} = \frac{4.2 \cdot 10^{2} \cdot 100}{1.15 \cdot 10^{3}}$$

$$k_{2,\ 22} = 36.5\ y^{-1}$$

Local atmosphere - local biota (22, 26)

The flux of carbon from the atmosphere to biota occurs mainly through primary production during daylight photosynthesis. An average flux of $4.9 \cdot 10^{3}$ g $ha^{-1}y^{-1}$, adopted by McKee and Rowsell (1984) was assumed to be valid for Swedish conditions too. The transfer coefficient is then

$$k_{22,\ 26} = \frac{100 \cdot 0.49}{4.21 \cdot 10^{8}}$$

$$k_{22,\ 26} = 1.6 \cdot 10^{-7}\ y^{-1}$$

Local biota - local atmosphere (26, 22)

Certain amounts of the carbon in living plants are returned to the atmosphere by respiration. The above-ground loss of CO_2 by autotrophic biota, expressed as a fraction of the gross primary production, is generally in the range 30 - 60 % (Westlake, 1963). Olson (1975) estimated a R/GPP quotient of 40 - 58 % and 74 - 79 % for temperate deciduous and cool temperature to boreal conifer forest, respectively. A value of 60 % was assumed to be valid for Swedish conditions. A GPP value of $1.17 \cdot 10^{7}$ g C $ha^{-1}y^{-1}$, assessed by Whittaker (1970) for a temperate forest, was applied, giving a respiration rate of $7.02 \cdot 10^{6}$ g C - $ha^{-1}y^{-1}$. 75 % of this value is supposed to constitute the above-ground loss to the atmosphere, while the remaining 25 % is respired through the roots (Harris et al., 1975). Thus, $5.27 \cdot 10^{6}$ g C $ha^{-1}y^{-1}$ is assumed to be lost from the biota compartment to the atmosphere. The transfer coefficient is

$$k = \frac{5.27 \cdot 10^{2} \cdot 100}{4.17 \cdot 10^{3}}$$

$$k = 12.64\ y^{-1}$$

Local biota - local surface soil (26, 2)

25 % of the respired carbon is lost from the biota through the roots (Harris et al., 1975). In accordance with the calculations for the local biota-local atmosphere transfer coefficient, $1.76 \cdot 10^{2}$ g C $m^{-2}y^{-1}$ is lost to the surface soil, meaning a transfer coefficient of

$$k_{26,\ 2} = \frac{1.76 \cdot 10^{2} \cdot 100}{4.17 \cdot 10^{3}}$$

$$k_{26,\ 2} = 4.22\ y^{-1}$$

Local atmosphere - regional biota (22, 27)

Using the same flux rate as for the local biota compartment, the atmosphere to biota transfer coefficient becomes

$$k_{22,\ 27} = \frac{1\cdot10^{5}\ \cdot\ 0.49}{4.21\cdot10^{8}}$$

$$k_{22,\ 27} = 1.16\cdot10^{-4}\ \ y^{-1}$$

Regional biota - local atmosphere (27, 22)

Adopting the same approach as for the local biota compartment, the transfer coefficient becomes

$$k_{27,\ 22} = \frac{5.27\cdot10^{2}\cdot1\cdot10^{5}}{6.55\cdot10^{6}}$$

$$k_{27,\ 22} = 8.05\ y^{-1}$$

Regional biota - regional surface soil (27, 19)

With the same assumptions as for the local biota, the transfer coefficient is

$$k_{27,\ 19} = \frac{1.76\cdot10^{2}\cdot1\cdot10^{5}}{6.55\cdot10^{6}}$$

$$k_{27,\ 19} = 2.69\ y^{-1}$$

Local atmosphere - regional surface soil (22, 19)

Adopting the atmosphere to soil flux rate given for the local surface soil, the transfer coefficient becomes

$$k_{22,\ 19} = \frac{1.9\cdot1\cdot10^{5}}{4.21\cdot10^{8}}$$

$$k_{22,\ 19} = 4.51\cdot10\ y^{-1}$$

Regional surface soil - local atmosphere (19, 22)

Using the same flux rate as for the local surface soil, the transfer coefficient becomes

$$k_{19,\ 22} = \frac{4.2 \cdot 10^{2} \cdot 1 \cdot 10^{5}}{1.1 \cdot 10^{9}}$$

$$k = 3.8 \cdot 10^{-2}\ y^{-1}$$

Regional surface soil - lake water (19, 3)

The average run-off in middle Sweden is about 7 l km^{-2} s^{-1} (Tryselius, 1971). The area of the surrounding drainage area is 17.5 km^2 and the total run-off is $1.07 \cdot 10^{6}$ l y^{-1}. Assuming a mean carbon content in run-off of 11 ppm, the fluvial input to the lake is $1.18 \cdot 10^{4}$ g C y^{-1}. The transfer coefficient surface soil-lake water is then

$$k_{19,\ 3} = \frac{1.18 \cdot 10^{4}}{1.93 \cdot 10^{11}}$$

$$k = 6.11 \cdot 10^{-8}\ y^{-1}$$

Local atmosphere - lake (22, 3)

For the SFR lake, a carbon exchange of 50 g C m^{-2} y^{-1} was assumed to occur in both directions through the air-water interface (McKee and Rowsell, 1984) giving a total influx of $1.25 \cdot 10^{8}$ g C y^{-1} to the lake system. The carbon inventory in the local atmospheric compartment is $4.21 \cdot 10^{8}$ g, and the transfer coefficient is

$$k_{22,\ 3} = \frac{1.25 \cdot 10^{8}}{4.21 \cdot 10^{8}}$$

$$k_{22,\ 3} = 2.97 \cdot 10^{-1}\ y^{-1}$$

Lake - local atmosphere (3, 22)

A net evasion of CO_2 from lake water to atmosphere of 24 and 150 g C $m^{-2}y^{-1}$ was estimated for two oligotrophic lakes, respectively (Lake 224, Hesslein et al., 1980; Lake 302, Bower and McCorkle, 1980). A corresponding value of 147 g C $m^{-2}y^{-1}$ was assessed for Lake Ontario by Robertson and Eadie (1975).

For the SFR lake, an assumed outflux of 50 g C m^{-2} y^{-1} will give a total loss of $1.25 \cdot 8$ g C y^{-1}. The carbon inventory in the lake is $8.25 \cdot 10^7$ g and the transfer rate is

$$k_{3,\ 22} = \frac{1.25 \cdot 10^8}{8.25 \cdot 10^7}$$

$$k_{3,\ 22} = 1.51\ y^{-1}$$

Fish - lake water (23,3)

Natural fish populations lose members by natural mortality, but also due to removal by the fishery. In long-lived species, the mortality rate seems to be effectively constant over a considerable age span at about 5 to 10 % per year (Beverton and Holt, 1959). Swedish data on perch (Alm, 1952) showed no evidence of a varying age-specific mortality rate. Of short-lived species, nearly all have a survival curve with some degree of downward curvature over nearly the whole range. This variation of the mortality rate with age reaches an extreme form in species where all or nearly all individuals die at, or soon after, spawning for the first time. Thus, the average mortality rate is not a particularly useful measure where the mortality rate is highly age-specific, but is satisfactory in the long-lived species with nearly linear survival curves. For the SFR lake, long-lived species such as perch and pike are assumed to make a major contribution to the fish biomass, and a mean transfer coefficient of 0.2 y^{-1} was chosen.

Lake water - fish (3, 23)

The transfer coefficient between water and fish was calculated as follows:

$$\frac{dIf}{dt} = k_{w,\ f} \cdot I_w - k_{f,\ w} \cdot I_f - \lambda \cdot I_f$$

where

$\frac{dIf}{dt}$ = Change in element inventory in the fish compartment

Iw; If = element inventories in water and fish, respectively

$k_{w,f}$; $k_{f,w}$ = transfer rates of element from water to fish, and vice versa (t^{-1}).

λ = radioactive decay (t^{-1})

At steady state $\frac{dIf}{dt} = 0$ and

$$k_{w,\ f} \cdot I_w = k_{f,\ w} \cdot I_f + \lambda \cdot I_f$$

$$k_{w,\ f} = \frac{If}{Iw} \cdot (kf,\ w + \lambda)$$

$\frac{If}{Iw}$ can be substituted by $\frac{Mf}{Mw} \cdot CF$

where

Mf; Mw = amounts of fish and water, respectively
CF = fish/water concentration factor

The transfer coefficient is then

$$k_{wf} = \frac{Mf}{Mw} \cdot CF\ (k_{f,\ w} + \lambda)$$

The standing stock of the lake fish is assumed to be 2 500 kg. The amount of lake water is $7.5 \cdot 10^9$ l and the fish/water concentration factor is 4 600 (Bergström and Puigdomenech, 1987). The transfer coefficient becomes

$$k_{3,\ 23} = \frac{2.5 \cdot 10^3}{7.5 \cdot 10^9} \cdot 4\ 600 \cdot 0.2$$

$$k_{3,\ 23} = 3.1 \cdot 10^{-4}\ y^{-1}$$

Lake water - lake surface sediment (3, 4)

Permanent sedimentation values ranged 10 - 15 g C $m^{-2}y^{-1}$ for oligotrophic and eutrophic conditions, respectively (Mirror Lake, Jordan and Likens, 1975; Lawrence Lake, Wetzel et al., 1972). Benthic respiration in oligotrophic lakes were ranging 20 - 80 g C $m^{-2}y^{-1}$ (Mirror Lake, Jordan and Likens, 1975; Lake 224 , Hesslein et al., 1980). For eutrophic systems, a value of 117 g C $m^{-2}y^{-1}$ was estimated in two lakes (Lawrence Lake, Wetzel et al., 1972; Lake 227, Hesslein, 1976). Resuspension seems to be of the same magnitude as the benthic output due to respiration. For the SFR lake, an assumed permanent sedimentation of 10 g C $m^{-2}y^{-1}$, a benthic respiration of 75 g C $m^{-2}y^{-1}$, and a resuspension of 75 g c $m^{-2}y^{-1}$ will give a total benthic input of 160 g C $m^{-2}y^{-1}$. A lake area of 2.5 km^2 will give rise to a gross carbon input to the lake sediment of $4.0 \cdot 10^8$ g C y^{-1}. The lake volume is $7.5 \cdot 10^9$ l and the mean carbon concentration is 11 ppm, giving an inventory of $8.25 \cdot 10^7$ g C. The transfer coefficient water-sediment is then

$$k = \frac{4.0 \cdot 10^8}{8.25 \cdot 10^7}$$

$$k_{3,\ 4} = 4.85\ y^{-1}$$

Lake surface sediment - lake water (4, 3)

Benthic respiration values in oligotrophic lakes were ranging 20 - 80 g C $m^{-2}y^{-1}$ (Mirror Lake, Jordan and Likens, 1975; Lake 224, Hesslein et al., 1980). For eutrophic systems, a value of 117 g C $m^{-2}y^{-1}$ was assessed in two lakes (Lawrence Lake, Wetzel et al., 1972; Lake 227, Hesslein, 1976). Besides, in Lawrence Lake, resuspension contributed an equal amount to be added to the benthic respiration figure. An assumed total output of 150 g C $m^{-2}y^{-1}$ from the sediment of the SFR lake, combined with the inventory of carbon in the surface sediment compartment (area 2.5 km^2, sediment depth 0.1 m, density 1.2, carbon content 5 % dw, water content 88 %) will give the transfer coefficient

$$k_{4,\,3} = \frac{3.75 \cdot 10^{8}}{3 \cdot 10^{9}}$$

$$k_{4,\,3} = 0.13\ y^{-1}$$

Lake surface sediment - lake deep sediment (4, 5)

An assumed permanent input of about 10 g C $m^{-2}y^{1}$ of sedimentary carbon will give a total benthic gain of $2.5 \cdot 10^{7}$ g C y^{-1}. The inventory of carbon in the surface sediment compartment is $3 \cdot 10^{9}$ g, and the transfer coefficient is

$$k_{4,\,5} = \frac{2.5 \cdot 10^{7}}{3 \cdot 10^{9}}$$

$$k_{4,\,5} = 8.3 \cdot 10^{-3}\ y^{-1}$$

Local atmosphere - regional atmosphere (22, 18)

In the SFR study (Bergström and Puigdomenech, 1987) the masses of the regional and global atmospheric compartments were $4.4 \cdot 10^{14}$ kg and $4.4 \cdot 10^{18}$ kg, respectively, and the transfer coefficient regional atmosphere-global atmosphere was 150 y^{-1}. The regional atmosphere covered an area of $3.7 \cdot 10^{5}$ km^{2}, meaning an exchange rate of $1.78 \cdot 10^{11}$ kg air per km^{2} and year. If a similar exchange rate can be adopted for the local atmospheric compartment, which covers an area of 2.6 km^{2}, the exchange is $4.63 \cdot 10^{11}$ kg y^{-1}, and the transfer coefficient local atmosphere-regional atmosphere should be of the order of $2 \cdot 10^{4}$ y^{-1}.

Regional atmosphere - local atmosphere (18, 22)

With the same approach as described above, the transfer coefficient regional atmosphere-local atmosphere becomes about $1y^{-1}$

Regional atmosphere - Baltic Sea (18, 6)

Using the flux rates through the air-water interface given by Nair (1981) for cold ocean conditions, and assuming a 25 % reduction to account for ice cover, an influx of 258 g C m^{-2} y^{-1} and an outflux of 158 g C m^{-2} y^{-1} was adopted for the Baltic Sea. A surface area of $3.7 \cdot 10^5$ km^2 will give a total annual influx of $9.55 \cdot 10^7$ tons of C y^{-1}. The inventory of carbon in the regional atmospheric compartment is $5.5 \cdot 10^7$ tons and the transfer coefficient atmosphere-water is then

$$k_{18,\ 6} = \frac{9.55 \cdot 10^7}{5.5 \cdot 10^7}$$

$$k_{18,\ 6} = 1.74\ y^{-1}$$

Baltic Sea - regional atmosphere (6, 18)

An outflux of 158 g C m^{-2} y^{-1} (see above) and an area of $3.7 \cdot 10^5$ km^2 will give a total loss of $5.85 \cdot 10^7$ tons C y^{-1} from the Baltic Sea water to the overlying atmosphere. The inventory of carbon in Baltic Sea water is $4.6 \cdot 10^8$ tons and the transfer coefficient becomes

$$k_{6,\ 18} = \frac{5.85 \cdot 10^7}{4.6 \cdot 10^8}$$

$$k_{6,\ 18} = 0.13\ y^{-1}$$

Fish - Baltic Sea (24, 6)

Using the same approach as for lake fish, a transfer coefficient of 0.2 y^{-1} was assumed to be valid.

Baltic Sea - fish (6, 24)

The same approach was used to calculate the water to fish transfer coefficient as for the lake fish. The volume of the Baltic Sea water amounts to 21 800 km³, and the fish biomass is about $4.1 \cdot 10^{6}$ tons (Thurow, 1980). The fish/water concentration factor is 4 600 (Bergström and Puigdomenech, 1987). The transfer coefficient is then

$$k_{6,\ 24} = \frac{4.1 \cdot 10^{6}}{2.18 \cdot 10^{13}} \cdot 4600 \cdot 0.2$$

$$k_{6,\ 24} = 1.7 \cdot 10^{-4}\ y^{-1}$$

Baltic Sea - Baltic Sea surface sediment (6, 7)

Elmgren (1982) estimated the total annual organic input to the benthic systems below 25 depth to be 50 to 60 g C m^{-2} in the Baltic Proper, about 40 g C m^{-2} in the Bothnian Sea, and about 20 g C m^{-2} in the Bothnian Bay. Adopting 40 g C m^{-2} y^{-1} as a mean value for the total area of $3.7 \cdot 10^{5}$ km², an annual input of $1.48 \cdot 10^{7}$ tons of C will occur to the sediments. An inventory of $4.6 \cdot 10^{8}$ tons of C in the Baltic Sea compartment will give a transfer coefficient of

$$k_{6,\ 7} = \frac{1.48 \cdot 10^{7}}{4.6 \cdot 10^{8}}$$

$$k_{6,\ 7} = 3.2 \cdot 10^{-2}\ y^{-1}$$

Baltic surface sediment-Baltic Sea (7, 6)

A total benthic respiration of about 50 g C m^{-2} y^{-1} was calculated by Elmgren (1982) for the Baltic proper and the Bothnian Sea. For a total area of $3.7 \cdot 10^{5}$ km², the benthic loss amounts to $1.85 \cdot 10^{7}$ tons C y^{-1}. The inventory of carbon in the surface sediment compartment is $1.8 \cdot 10^{8}$ tons and the transfer coefficient is

$$k_{7,\ 6} = \frac{1.85 \cdot 10^{7}}{1.8 \cdot 10^{8}}$$

$$k_{7,\ 6} = 0.10\ y^{-1}$$

Baltic Sea surface sediment - Baltic Sea deep sediment (7, 8)

It is assumed that 10 g C m^{-2} y^{-1} will be buried annually in the Baltic Sediments. For an area of $3.7 \cdot 10^{5}$ km^{2}, a total input of $3.7 \cdot 10^{6}$ tons of C y^{-1} will occur to the deep sediment compartment. The inventory of carbon in the surface sediment is $1.8 \cdot 10^{8}$ tons, and the transfer coefficient is then

$$k_{7,\ 8} = \frac{3.7 \cdot 10^{6}}{1.8 \cdot 10^{8}}$$

$$k_{7,\ 8} = 2.1 \cdot 10^{-2}\ y^{-1}$$

Global atmosphere - global sea surface (17, 9)

There is a great deal of conflicting literature concerning the large-scale exchange of carbon between the atmosphere and the ocean. The abundance of C-14 in the atmosphere showed a sharp increase during the late 1950's and early 1960's, reflecting the period of active bomb testing. After 1963, the $^{14}CO_2$ content of the atmosphere decreased, implying a transfer to other reservoirs. The mean residence time in the atmosphere for CO_2 is about 5 years, and Machta (1973) suggested that the atmospheric decrease was a result of CO_2 entering the ocean. Other authors also assume net flux into the ocean (e.g. Broecker et al., 1979; Nair (1981). However, as discussed by, e.g. Nair (1981), CO_2 dissolved in sea-water is the only component of the carbonate system in direct exchange with the atmospheric CO_2. The amount of dissolved CO_2 constitutes only a minor part of the total DIC pool. If the transfer of carbon from the atmosphere to ocean water is instantaneously increased, the CO_2 partial pressure will increase much more rapidly than the corresponding total DIC inventory. The increased CO_2

partial pressure results in an enhanced reverse flux to the atmosphere. Thus, increased CO_2 partial pressure will produce only a small increase in the total DIC pool, and the oceans would be much less of a sink for CO_2 than would be expected.

The flux of carbon from the atmosphere to the ocean surface ranges 190 - 340 g C $m^{-2}y^{-1}$ (Broecker et al., 1979; Peng et al., 1979; Nair, 1981, Emanuel et al., 1981). The estimated carbon inventory of the global atmospheric compartment ranges 550 - 631 GT (Killough, 1980; Hoffert et al., 1981; Nair, 1981; this study). Taking into consideration the variation in estimated compartment inventories and flux rates, a mean value of 0.05 y^{-1} was adopted in this study.

Global sea surface - global atmosphere (9, 17)

A transfer rate of 0.1 y^{-1}, used by UNSCEAR (1982) was used.

Global sea surface - deep sea (9, 12)

Bergman and McEwan (1977) estimated fractional transfer rates between $5 \cdot 10^{-2}$ and $1.7 \cdot 10^{-1}$ y^{-1} for different geographical zones, while Emanuel et al. (1984) estimated a mean value of $1.30 \cdot 10^{-2}$ y^{-1}. The value $1 \cdot 10^{-1}$ y^{-1}, used by Bergström and Puigdomenech (1987), was assumed to be valid.

Deep sea - global sea surface (12, 9)

The mean residence time for carbon in the deep sea region is about 1000 y (Stuiver, 1973). A transfer coefficient of $2 \cdot 10^{-3}$ y^{-1} was adopted.

Global fish - global sea surface (25, 9)

Applying a mean mortality rate which includes the global fish stock will by necessity involve a high degree of uncertainty. However, in conformity with the lake and the Baltic Sea, a mean transfer coefficient of 0.2 y^{-1} was chosen in this study.

Global sea surface - global fish (9, 25)

A global fish production of $2.4 \cdot 10^8$ tons y^{-1} was assessed by Ryther (1969). A mean production/biomass ratio of $0.2\ y^{-1}$ will give a fish stock of $1.2 \cdot 10^9$ tons. The transfer coefficient was calculated using the same approach as for the lake and Baltic Sea fish populations. The mass of water of the global sea surface compartment is $6.1 \cdot 10^{16}\ m^3$, and the fish/water concentration factor is 1 800 (Bergström and Puigdomenech, 1987).

$$k_{9,\ 25} = \frac{1.2 \cdot 10^9}{6.1 \cdot 10^{16}} \cdot 1\ 800 \cdot 0.2$$

$$k_{9,\ 25} = 7.1 \cdot 10^{-6}\ y^{-1}$$

Global sea - surface sediment (9, 10)

About $15 \cdot 10^9$ tons of carbon are removed from the surface of the open ocean each year via the sinking of POC (Jenkins, 1982; Jenkins and Goldman, 1985; Martin et al., 1987). However, much of this carbon is regenerated before the particles reach the ocean floor. About 50 % of this carbon removed from the surface is regenerated at depths less than 300 m (Martin et al., 1987). The inventory of carbon in the global sea surface compartment is $1.5 \cdot 10^{12}$ tons (Killough, 1980). The sea-sediment transfer coefficient will then be

$$k_{9,\ 10} = \frac{7.5 \cdot 10^9}{1.5 \cdot 10^{12}}$$

$$k_{9,\ 10} = 5.10^{-3}\ y^{-1}$$

Marine surface sediment - global sea surface (10, 9)

In analogy with the conditions in the Baltic Sea, a transfer coefficient of $0.10\ y^{-1}$ was applied to the fractional transfer between the shelf sediment and the overlying water column.

Marine surface sediment - marine deep sediment (10, 11)

In consistency with the conditions in the Baltic Sea, a transfer coefficient of $2.1 \cdot 10^{-2}$ y^{-1} was assumed to be also valid for shelf conditions.

Global atmosphere - global soil (17, 15)

A transfer coefficient of 0.1 y^{-1} was used by UNSCEAR (1982) and was also adopted in this study.

Global soil - global atmosphere (15, 17)

The fractional transfer of $1 \cdot 10^{-2}$ y^{-1}, estimated by Bergman and McEwan (1977), was adopted in this study.

Global atmosphere - global long-lived biota (17, 28)

The global terrestrial net production of long-lived biota is $4.9 \cdot 10^{10}$ tons of carbon y^{-1} (Woodwell et al., 1987). An atmospheric carbon inventory of $5.5 \cdot 10^{11}$ tons will give a transfer coefficient of

$$k_{17,\,28} = \frac{4.9 \cdot 10^{10}}{5.5 \cdot 10^{11}}$$

$$k_{17,\,28} = 8.9 \cdot 10^{-2}\ y^{-1}$$

Global long lived biota - global atmosphere (28, 17)

The global plant area of carbon constitutes $8.3 \cdot 10^{11}$ tons, and the yearly release from this pool amounts to $7.8 \cdot 10^{9}$ tons (Woodwell et al., 1987). The transfer coefficient becomes

$$k_{28,\,17} = \frac{7.8 \cdot 10^{9}}{8.3 \cdot 10^{11}}$$

$$k_{28,\,17} = 9.4 \cdot 10^{-3}\ y^{-1}$$

This is almost identical to the estimated residence time of 100 y^{-1} which will give a transfer coefficient of $1 \cdot 10^{-2}$ y^{-1}.

Global atmosphere - global short-lived biota (17, 29)

The global net production from cultivated land amounts to $4.2 \cdot 10^{9}$ tons of carbon y^{-1} (Woodwell et al., 1978). An atmospheric carbon inventory of $5.5 \cdot 10^{11}$ tons gives a transfer coefficient of

$$k_{17,\ 29} = \frac{4.2 \cdot 10^{9}}{5.5 \cdot 10^{11}}$$

$$k_{17,\ 29} = 7.6 \cdot 10^{-3}\ y^{-1}$$

Global short lived biota - global atmosphere (29, 17)

A mean carbon residence time of 2 y will give a transfer coefficient of

$$k_{29,\ 17} = 0.5\ y^{-1}$$

Global long-lived biota - global soil (28, 15)

The total plant mass of carbon constitutes $8.3 \cdot 10^{11}$ tons and the total net production amounts to $5.3 \cdot 10^{10}$ tons (Woodwell et al., 1978). Gross primary production is derived by adding respiration to the net production value. Assuming that 60 % of the GPP constitutes respiration (Westlake, 1963), the world GPP amounts to $1.32 \cdot 10^{11}$-tons of carbon. With the same assumptions as for the local biota-local surface soil transfer coefficient, the below-ground loss of carbon via the roots is $1.98 \cdot 10^{10}$ tons of carbon y^{-1}. The corresponding transfer coefficient is then

$$k_{28,\ 15} = \frac{1.98 \cdot 10^{10}}{8.3 \cdot 10^{11}}$$

$$k_{28,\ 15} = 2.39 \cdot 10^{-2}$$

Global short-lived biota - global soil (29, 15)

In agreement with the long-lived biota, the transfer coefficient is

$$k_{29,\ 15} = 2.4 \cdot 10^{-2}\ y^{-1}$$

Global groundwater - global sea surface (16, 9)

A transfer rate of $2.9 \cdot 10^{-3}$ y^{-1} was used to assess the transfer from global groundwater to global soil. The same value was chosen for the global groundwater-global sea surface transfer rate.

For the remaining transfer coefficients, the data by Bergström and Puigdomenech (1987) were used. All transfer coefficients are compiled in Table 17.

Table 17

Transfer coefficients (y^{-1}) used in this study

		mean value	range	distribution
local groundwater - local surface soil	1,2	4.4E-4		
local groundwater - lake water	1,3	1.0E0		
local surface soil - local deep soil	2,14	9.1E-1		
local surface soil - local atmosphere	2,22	3.65E1		
lake water - lake surface sediment	3,4	4.85E0		
lake water - Baltic Sea water	3,6	1.0E0		
lake water - regional surface soil	3,19	3.0E-3		
lake water - local atmosphere	3,22	1.5E0		
lake water - lake fish	3,23	3.1E-4		
Lake surface sediment - lake water	4,3	1.3E-1		
lake surface sediment - lake deep sediment	4,5	8.3E-3		
Baltic Sea water - Baltic surface sediment	6,7	3.2E-2		
Baltic Sea water - global sea surface	6,9	4.3E-2		
Baltic Sea water - regional atmosphere	6,18	1.3E-1		
Baltic Sea water - Baltic fish	6,24	1.7E-4		
Baltic surface sediment - Baltic Sea water	7,6	1.0E-1		
Baltic surface sediment - Baltic deep sediment	7,8	2.1E-2		
global sea surface - Baltic Sea water	9,6	4.7E-5		

Table 17 (Cont'd)

		mean value	range	distribution
global sea surface - marine surface sediment	9,10	5.0E-3		
global sea surface - global deep sea	9,12	1.0E-1		
global sea surface - global soil	9,15	1.7E-7		
global sea surface - global atmosphere	9,17	5.0E-1		
global sea surface - global fish	9,25	7.1E-6		
marine surface sediment - global sea surface	10,9	1.0E-1		
marine surface sediment - marine deep sediment	10,11	2.1E-2		
global deep sea - global sea surface	12,9	2.0E-3		
global deep sea - deep sea sediment	12,13	1.0E-7		
local deep soil - local groundwater	14,1	1.8E-1		
global soil - global sea surface	15,9	4.4E-1		
global soil - global groundwater	15,16	3.8E-1		
global soil - global atmosphere	15,17	1.0E-2		
global groundwater - global sea surface	16,9	2.9E-3		
global groundwater - global soil	16,15	2.9E-3		
global atmosphere - global sea surface	17,9	5.0E-2		

Table 17 (Cont'd)

		mean value	range	distribution
global atmosphere - global soil	17,15	1.0E-1		
global atmosphere - regional atmosphere	17,18	1.6E-2		
global atmosphere - global long-lived biota	17,28	8.9E-2		
global atmosphere - global short-lived biota	17,29	7.6E-3		
regional atmosphere - Baltic Sea	18,6	1.74EO		
regional atmosphere - global atmosphere	18,17	150EO		
regional atmosphere - local atmosphere	18,22	1.0E0		
regional surface soil - lake water	19,3	6.1E-8		
regional surface soil - regional deep soil	19,20	9.1E-1		
regional surface soil - local atmosphere	19,22	3.8E-2		
regional deep soil - regional groundwater	20,21	1.8EO		
regional groundwater - lake water	21,3	2.0E-1		
local atmosphere - local surface soil	22,2	4.5E-7		
local atmosphere - lake water	22,3	2.9E-1		
local atmosphere - regional atmosphere	22,18	2.0E4		

Table 17 (cont'd)

		mean value	range	distribution
local atmosphere - regional surface soil	22,19	4.5E-4		
local atmosphere - local biota	22,26	1.1E-7		
local atmosphere - regional biota	22,27	1.1E-4		
lake fish - lake water	23,3	2.0E-1		
Baltic fish - Baltic Sea water	24,6	2.0E-1		
global fish - global sea surface	25,9	2.0E-1		
local biota - local surface soil	26,2	4.2EO		
local biota - local atmosphere	26,22	1.26E1		
regional biota - regional surface soil	27,19	2.7EO		
regional biota - local atmosphere	27,22	8.0EO		
global biota - global soil	28,15	2.4E-2		
global long-lived biota - global atmosphere	28,17	9.4E-3		
global short-lived biota - global soil	29,15	2.4E-2		
global short-lived biota - global atmosphere	29,17	5.0E-1		

5 Dose conversion factor

In ICRP Publication 30, Part 3, the fractional absorption to blood of dietary carbon in the form of organic compounds labelled with carbon-14 is taken to be 1 and it is assumed that inhaled or ingested C-14 labelled compounds are instantaneously uniformly distributed throughout all organs and tissues of the body where they are retained with a biological half-life of 40 days (ICRP, 1978). It is considered that this assumption will yield realistic whole body doses for C-14 labelled metabolites and that it will overestimate whole body doses from most other C-14 labelled compounds. It is further assumed that all carbon dioxide entering the respiratory system is translocated to blood. Carbon dioxide in the blood exists mainly as bicarbonate.

It is assumed that carbon inhaled as carbon dioxide is uniformly distributed throughout all organs and tissues of the body and that its retention is governed by the retention function.

$$R(t) = 0.18\ e^{-0.693t/5} + 0.81\ e^{-0.693t/60} + 0.01\ e^{-0.693t/60000}$$

where t is in minutes and 60 000 min is the biological half-life for dietary carbon as estimated from Reference Man (ICRP, 1975). This 60 000 min half-life is based on the assumption that the small fraction of carbon incorporated into tissues following inhalation of CO_2 will behave in a similar fashion to carbon entering the systematic circulation following ingestion of dietary carbon.

These assumptions lead to the following annual limits on intake (ALI):

Annual limits on intake, ALI(Bq) and derived air concentrations, DAC(BQ m^{-3}) (40 h wk) for isotopes of carbon.

Labelled organic compounds

Radionuclide		Oral	Inhalation
C-14	ALI	$9 \cdot 10^7$	$9 \cdot 10^7$
	DAC	-	$4 \cdot 10^4$

Carbon dioxide

Radionuclide		Inhalation
C-14	ALI	$8 \cdot 10^9$
	DAC	$3 \cdot 10^6$

Annual limits on intake by inhalation of carbon dioxide are about 90 (89) times the figure for inhalation of labelled organic compounds. No figure is given for oral intake of labelled inorganic compounds, e.g. bicarbonate in drinking water. The annual limits on intake of labelled organic compounds are, however, the same for oral intake and for inhalation. Is it too bold an assumption to consider that annual limits on oral intake of labelled inorganic compounds, e.g. bicarbonate, should be the same as on inhalation of labelled carbon dioxide?

If this assumption is correct, the ALI value for oral intake of labelled carbonic acid, bicarbonate and carbonate would be 8E09 Bq (instead of 9E07 Bq).

The fact that C-14 metabolism and toxicity depend on the form of the chemical compound metabolized is stressed in some French, Russian and Canadian studies (Jeanmaire, 1982; Vasilenko et al., 1979; Whillans and Johnson, 1985). Inorganic C-14 compounds are metabolized quickly. A rapid excretion of most of the absorbed fraction is observed. The retention time of C-14 organic compounds in the body is much longer (Simmons et al., 1982).

In a Russian study (Osipov, 1982) it was noted that during administration of C-14 organic compounds the irradiation doses affecting organs and tissues were ten times higher than the doses from its inorganic compounds. This study would thus support a ten times higher ALI-value, i.e. 9E08 Bq, for ingestion of inorganic labelled compounds.

The suggested alteration of the ALI-values has implications for the dose factor. The sum of the weighted commited dose equivalent will be a factor of ten lower for the inorganic than for the organic labelled compound. Consequently, different dose conversion factors should be used for different modes of intake of ^{14}C, e.g. consumption of drinking water (mainly inorganic carbon) or different food products (mainly organic carbon).

6 Results and discussion

The well/lake scenario for ^{14}C released from the Swedish SFR site, performed by Bergström and Puigdomenech (1987), was re-evaluated. In their calculations, 10 % of the total inventory of ^{14}C in the repository was assumed to be unaffected by sorption. This simple assumption is somewhat incongruous to use in conjunction with model predictions based on a rather comprehensive review of carbon data. In a new set of release data for ^{14}C, supplied by Puigdomenech in the second part of this report, the total inventory of ^{14}C was released from the repository and underwent dispersion and retardation in the geological media before entering the biosphere.

The individual doses for the local, regional and intermediate zones peaked at about $2.4 \cdot 10^4$ y after repository closure with $5 \cdot 10^{-3}$, $8 \cdot 10^{-4}$ and $2 \cdot 10^{-9}$ µSv y^{-1}, respectively. The individual global and collective doses reached a maximum of $4 \cdot 10^{-9}$ µSv y^{-1} and $4 \cdot 10^{-5}$ manSv y^{-1}, respectively, at $3.1 \cdot 10^4$ y. A total release of 8.4 Gbq of ^{14}C will yield a total collective dose commitment of 1.1 manSv or 130 manSv Tbq^{-1}.

Vegetables and water consumption contributed 41 and 37 %, respectively, of the annual individual dose in the local zone. For the regional and intermediate areas, fish consumption contributed 99 and 79 %, respectively. The latter results are similar to those presented earlier: the discharge of ^{14}C from the SFR repository into lake water and the subsequent exposure to edible fish will result in the most serious health consequences, in relation to other exposure routes. This is also in accordance with the findings by Bush et al (1984); if the discharge of ^{14}C occurred to a freshwater recipient, about half the collective dose commitment would arise from the ingestion of drinking water and half from the ingestion of fish.

In the global model, the assumption of rapid mixing is invalid in the short term perspective and calls for a range of regional models. This will justify the approach by Bergström and Puigdomenech (1987). However, owing to the long half-life of ^{14}C compared with the environmental flux rates of carbon, the collective dose commitment over infinite time is not affected by the manner in which ^{14}C is discharged, apart from some differences in the first few centuries. Estimates of the dose commitment are also fairly insensitive to the degree of resolution of the model. In the global model, ^{14}C enters the natural global carbon cycle and mixes rapidly with stable carbon to establish a uniform specific activity throughout large reservoirs. In a long-term perspective, all global ^{14}C models tend towards agreement, since the ultimate distribution of ^{14}C is largely affected by the sizes of the major carbon reservoirs, i.e., the atmosphere and the deep sea, and by a few slow transfer processes.

The global dose commitment derived from ^{14}C releases from the nuclear industry is estimated by modelling the environmental distribution and behaviour of naturally produced ^{14}C. In an attempt to check the reliability of the model, a simulation was performed in which an amount of ^{14}C as large as the yearly production of ^{14}C due to cosmic radiation was released into the global atmospheric compartment. The calculated ^{14}C concentration in global biota at steady state was 173 Bq kg^{-1}, yielding a specific activity of 333 Bq/kg C (compartment mass $1.6 \cdot 10^{15}$ kg, carbon inventory $8.3 \cdot 10^{14}$ kg). This is somewhat higher than the value of 227 Bq/kg C given by UNSCEAR (1982). The global individual dose arising from the exposure of naturally produced ^{14}C was 26 µSv y^{-1}, which can be compared with the corresponding value of 12 µSv y^{-1} reported by UNSCEAR.

The estimated global collective dose commitment due to the release of ^{14}C from the SFR site was 130 manSv TBq^{-1} which can be compared with the corresponding values 70, 120, 67 and 100 manSv TBq^{-1} obtained by Bergman and McEwan (1977), Killough (1980), UNSCEAR (1982) and Bush et al. (1984), respectively. Accordingly, the model seems to predict the global turnover of carbon adequately, at least in comparison with earlier results.

The evolution of methane in the hypothetical SFR lake may be predicted based on the preceding data on methane release rates and bubble formation in similar freshwater systems. A predicted net sedimentation rate of 3 mm y^{-1} yields a permanent deposition of about 36 g C $m^{-2}y^{-1}$ to the lake bottom, assuming a water content of 80 %, a density of 1.2, and an organic carbon content of 5 % dw in the sedimenting material.

According to the in-situ measurements of methane release versus sedimentation (Figure 4), a carbon input of 36 g C $m^{-2}y^{-1}$ will generate about 10 - 20 g CH_4-C $m^{-2}y^{-1}$. Of this amount, about 2 - 10 g CH_4-C $m^{-2}y^{-1}$ will be lost from the lake due to release to the atmosphere. By comparison, if ebullition constitutes 30 % of the total methane release, and 90 % of the bubbles escape to the atmosphere (Kelly and Chynoweth, 1979), about 5 g CH_4-C $m^{-2}y^{-1}$ will be lost. A methane production/sedimentation ratio of 27 %, also estimated by Kelly and Chynoweth (1979), will give a release of methane to the atmosphere of about 3 g C $m^{-2}y^{-1}$. These flux rates can be compared with an estimated flow of CO_2 from the lake sediment to the overlying water of 150 g C $m^{-2}y^{-1}$, and from the lake water to the atmosphere of 50 g C $m^{-2}y^{-1}$. Consequently, this route will only make a minor contribution to the overall loss of carbon from the lake ecosystem.

General conclusions

The initial amount of C-14 in the repository is assumed to be 5 TBq, which is close to the amount expected for the Swedish production by the year 2010 after it has decayed for 2500 years. The content of C-14 in spent ion exchange resins from PWR and BWR has been estimated to be in the ranges 2-24 and 2-132 GBq GW(e)$^{-1}$ y^{-1}, respectively. The scarcity and scatter in available data calls for further experimental efforts.

C-14 in anion exchange resins is expected to be present as $^{14}CO_3^{2-}$ species. The pore water in the concrete repository is expected to maintain an average pH value of >10.5 for a period of at least 10^6 years. The inorganic carbon concentration is likely to be controlled by $CaCO_3$ saturation. Any addition of carbonate will cause the precipitation of $CaCO_3$. The cement matrix of the repository will retain most of the 134 g of $^{14}CO_3^{2-}$ initially present.

Bacterial production of CO_2 and CH_4 from the degradation of ion-exchange resins and bitumen might contribute to ^{14}C release to the biosphere. However, there are large uncertainties in the release of ^{14}C associated to microbial reactions. Methane contributes only to a small extent to the overall loss of carbon from the freshwater ecosystem. High pH values expected in the cement repository might reduce the rate of microbial gas production by some orders of magnitude.

The dose response from ^{14}C releases is assessed by the use of a compartment model which reflects the environmental distribution and behaviour of naturally-produced ^{14}C. The indivdual doses for the local, regional and intermediate zones peaked with $5 \cdot 10^{-3}$, $8 \cdot 10^{-4}$, and $2 \cdot 10^{-9}$ µSv y^{-1}, respectively, at about 24 000 years. The individual global and collective doses reached a maximum of $4 \cdot 10^{-9}$ µSv y^{-1} and $4 \cdot 10^{-5}$ manSv y^{-1}, respectively, at 38 000 years. A total release of 8.4 GBq of ^{14}C will yield a total collective dose commitment of 1.1 manSv or 130 manSv TBq^{1-}, which is in concordance with earlier findings (Bergman and McEwan, 1977; Killough, 1980; UNSCEAR, 1982; Bush et al.,1984). If ^{14}C from the SFR repository leaks into lake water the dominant exposure pathway is by consumption of edible fish.

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[1] Starprog AB
[2] SKB AB
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