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GLOBAL AND LOCAL EFFECTS OF ^{14}C DISCHARGES FROM THE NUCLEAR FUEL CYCLE

MARTIN McCARTNEY, M S BAXTER, KEITH McKAY
Department of Chemistry, University of Glasgow, G12 8QQ, Scotland

and

E MARIAN SCOTT
Department of Statistics, University of Glasgow

ABSTRACT. The radiologic impact of ^{14}C produced by the nuclear fuel cycle is assessed at both global and local levels. In the former context, it is predicted here that the specific activity of atmospheric CO_2 in the year 2050 will be ca 7.6 $pCig^{-1}C$. Although this is similar to the present level, the subsequent collective dose commitment could be highly significant.

The enhancement of ^{14}C concentrations around the nuclear fuel-reprocessing plant at Sellafield (Windscale) in Cumbria, U K has been monitored over recent years. For example, maximum levels of 27.2 $pCig^{-1}C$ (~350% above natural) during 1984 were observed <1km from the plant, with enhanced activities detectable to at least 29km. Nevertheless, it is clear that the radiologic significance to the local population is low. The spatial distribution of the excess ^{14}C allows atmospheric dispersion models to be tested in the context of continuous releases and the results thus far show that the Gaussian plume model performs successfully.

INTRODUCTION

^{14}C is produced naturally in the upper atmosphere at a rate which sustains a mean specific activity of 6.11 $pCig^{-1}C$ in both the atmosphere and terrestrial biosphere (Karlen *et al,* 1964). It is well known that deviations from this value have occurred in the past as a result of both natural (de Vries, 1958; Damon, Long & Grey, 1966; Suess & Stuiver, 1966) and anthropogenic (Suess, 1955; Houtermans, Suess & Munk, 1967; Nydal, 1968; Young & Fairhall, 1968) perturbations to the global carbon cycle. The most recent artificial modulation has been caused by the production of ^{14}C in nuclear reactors by a variety of mechanisms but prinicipally via the $^{14}N(n,p)^{14}C$, $^{13}C(n,\gamma)^{14}C$ and $^{17}O(n,\alpha)^{14}C$ reactions (Magno, Nelson & Ellet, 1974; Bush, White & Smith, 1983).

The environmental release of reactor-derived ^{14}C leads to an increase in atmospheric specific activity and, hence, to a concomitant increase in the radiation dose to man. It is to be expected that despite ecologic misgivings and to help meet the world's growing energy demand, installed nuclear energy capacity will continue to increase during the next century (Hafele, 1982). Thus, it is important to define the extent to which future ^{14}C levels might be perturbed and to assess the potential risk, if any, to the public from fuel cycle ^{14}C discharges. The objects of this study are: 1) to determine whether or not ^{14}C releases from the nuclear industry could lead to globally significant ^{14}C levels in the future, 2) to estimate the resulting population dose and, 3) at local level, to quantify and model the extent of the short-range enhancement of ^{14}C activities which can occur around large nuclear installations. This project is performed within the framework of an IAEA coordinated research program, on ^{14}C from nuclear installations, which is currently reviewing the impact of ^{14}C releases on present and future populations.

GLOBAL EFFECTS OF ^{14}C DISCHARGES

To estimate the future individual dose and collective dose commitment to man from ^{14}C emissions, specific activities of atmospheric CO_2 must first be predicted. Such calculations involve establishment of mathematical representations of the global carbon cycle and employment of future energy-economic models. The predictions of the latter can then be superimposed on to the carbon cycle models to produce atmospheric activity estimates.

There is, in fact, an extensive literature on carbon cycle models, these having been developed and applied for a wide range of purposes, eg, assessment of climatic change, Suess effect, weapons-derived ^{14}C, atmospheric CO_2 levels, etc. The aim of this study was to collate, adapt and use a frontier cross-section of these models for the singular purpose of predicting future atmospheric specific activities of, and exposures to, ^{14}C resulting from nuclear energy production. Towards this end, five carbon cycle models of varying complexity have been used thus far. The simplest is a 1-box model which merely defines the mean effective residence time of a CO_2 molecule in the atmosphere (Thommeret, Thommeret & Baxter, 1983). The 3-box model (Baxter & Walton, 1970) introduces the concept of exchange between the atmosphere, surface ocean/biosphere and deep ocean and this process is expanded in the 8-box model (Bush, White & Smith, 1983). In the 16-box model (Killough, 1980), the deep ocean is split into 12 compartments in an attempt to imitate diffusional transfer by the approach pioneered by Oeschger *et al* (1975). This technique is also incorporated in the 25-box model (Emanuel *et al,* 1984), which, in addition, contains a more detailed representation of the biosphere. The models were initially transferred to computer in Glasgow and then were tested and fine-tuned both in the natural steady-state mode and in their reproduction of the known anthropogenic trends in atmospheric specific activity over the past century. It was found here that several of the models were, in their published forms, unable to replicate natural ^{14}C equilibrium over long-term runs and the controlling parameters were subsequently adjusted slightly to maintain balance within the cycle.

Predictions of ^{14}C levels have been limited here to the period up to 2050 by the uncertainties inherent in future energy scenarios. Five different projections have been assessed to ensure coverage of the wide range of possible alternatives. Thus, two scenarios, generated by IIASA (Hafele, 1982) and catering for either a high (1) or low (2) global energy demand up to 2030, have been used. In addition, a standard case for the future growth of the nuclear industry has been defined (Edmonds & Reilly, 1983) and used in conjunction with 3 different scenarios (probable (4), high (5) and low (3)) for the future release of CO_2 from fossil fuel combustion (Edmonds *et al,* 1984). These projections of the installed nuclear energy capacity are combined with literature data on ^{14}C releases (Bush, White, & Smith, 1983) in Table 1 (which suggest a mean release rate of 47 Ci/GW(e) year) to generate annual figures for future ^{14}C discharges. Future atmospheric specific activities can be calculated, by feeding this data set into the carbon cycle models. It is worth noting here that our approach involves input of the

TABLE 1
Summary of ^{14}C waste arisings (Ci/GW(e) year)

Reactor type	Distribution (%)	Total production	Gaseous wastes	
			Reactor	Reprocessing plant
PWR	65	70	5	20
BWR	22	95	10	20
AGR	1	220	15	10
MAGNOX	1	380	30	100
LMFBR	1	25	0.02	5
HTGR	2	151	1	150
HWR	4	275	200	40
CANDU	4	275	200	40

same data sets to all models, thus eliminating previous result variability from this source.

For short-term studies (*ie*, up to 2050) the degree of inter-model deviation is small because for such time periods all model predictions are primarily governed by the exchange rate between the atmosphere and the surface ocean. This latter parameter is relatively well defined and hence all models yield similar results which can therefore be viewed with considerable confidence. The representative trends are shown in Figure 1 based on the simple 1-box model. Both IIASA scenarios (1) and (2) and hypothesis (4) produce virtually identical results. Thus, a value of 7.6 $pCig^{-1}C$ is predicted for the specific activity of atmospheric carbon in the year 2050 (all models give values in the range 7.0–7.6 $pCig^{-1}C$). Although this activity is nearly 25% above the natural baseline value, it is, of course, very similar to present levels. This finding reflects the competing nature of ^{14}C releases from the nuclear industry with inactive CO_2 releases from fossil fuel combustion. If, however, the future rate of fossil fuel combustion follows the low (3) or high (5) scenarios, activities of 8.5 and 5.6 $pCig^{-1}C$, respectively,

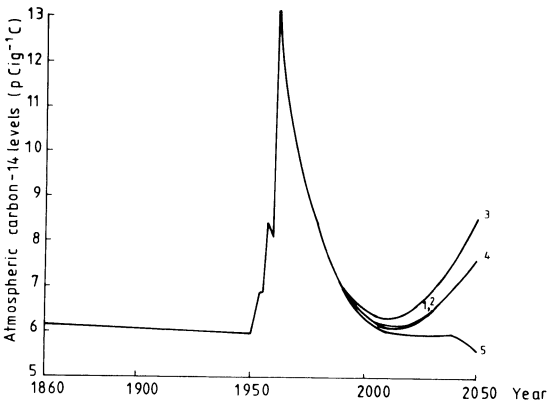

Fig 1. Predictions of atmospheric ^{14}C levels up to 2050

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would be expected in 2050. It has, in passing, also been shown that, although the effect of forest clearance on the carbon cycle is important from the climatic viewpoint, it has a negligible influence (<1.5%) on atmospheric specific activities. It is further possible that, in future, a major share of the nuclear energy market will gradually be transferred to fast breeder reactors and, since these systems have a very low ^{14}C production rate, this trend would lead to a corresponding reduction in ambient ^{14}C levels. Based on the most probable energy scenarios, however, it can be predicted that, by 2050, mean atmospheric ^{14}C activities will not significantly exceed present values and are likely to be in the range 7.5 ± 2 pCig^{-1}C.

On the above basis, however, it would be misleading to imply that, although individual exposures to ^{14}C will in the near future be low, global ^{14}C emissions will also be of negligible importance. The real and major significance of ^{14}C discharges is that, because of its long half-life (5730 years), initial gaseous form, direct incorporation into the biosphere and relatively high inventory in man, emissions of this nuclide will lead to relatively large exposures of global populations over very long time scales (Schwiback, Riedel & Bretschneider, 1978). This latter concept is incorporated in, and quantified by, the radiologic quantity, the collective dose commitment, which is a summation of all doses, however small, incurred by all people both at present and in the future (Beninson & Gonzalez, 1982). Essentially, it is derived by double integration of the population spectrum of individual dose rates in both space and time, the limits of integration being from zero to maximum doses and from time of release to infinity. The collective dose commitment is, therefore, a measure of the entire detriment resulting, in this case, from ^{14}C discharges. There are several important considerations: 1) it is currently believed that, of all nuclides discharged in routine operations by the nuclear industry, ^{14}C is likely to produce one of the largest collective dose commitments (Kelly *et al,* 1975; UNSCEAR, 1982); 2) in the process of optimizing nuclear waste control procedures, as recommended by ICRP (1976), collective dose commitment calculations now form an integral part; basically, collective dose commitments must be kept "as low as is reasonably achievable, economic and social considerations being taken into account" (ICRP, 1973, 1976). Thus, they must be calculated for each of a full range of possible disposal options. Studies of the temporal delivery of this dose and of its variation between alternative waste-processing and disposal strategies are, therefore, both of major interest and obligatory; 3) being central to the above optimization procedure, collective dose commitments should not take into account competing effects (eg, Suess effect) since, in this case, reactor-derived ^{14}C leads to an independent detriment. As a result, collective dose commitments are best estimated for unit environmental release of ^{14}C.

In this study the principal objective of the global box-modeling aspect is to assess the collective dose commitment resulting from 1 Ci release of ^{14}C and to investigate the likely variations in dose delivery, 1) with time and 2) between disposal options. The dose for any particular year can be calculated by combining the excess value (over the baseline level) of the atmospheric CO_2 specific activity with the dose rate factor of 2.08×10^6 Sv/yr

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per $Cig^{-1}C$ (Killough & Rohwer, 1978) and the population figure (taken to be a constant 10^{10}). The collective dose commitment is the summation of this figure from the year of release to infinity.

The results of this study (Fig 2) demonstrate how the collective dose commitment for a 1 Ci atmospheric release accumulates over time. Because of the need to integrate over long periods in this work, the more detailed carbon cycle models must be used and the results for the 3, 8, 16 and 25-box models are 4.2, 4.0, 5.6 and 4.4 man Sv, respectively. Once again, the aim in using a range of different carbon cycle models is to check the degree of inter-model correlation when used for a single data set and, hence, to indicate the variability which can be associated with the final result. An average value here of 4.6 ± 1.0 man Sv/Ci is in excellent agreement with other published data (Schwibach, Riedel & Bretschneider, 1978; Killough, 1980), with ca 8, 24, and 75% of the total dose being delivered in 100, 1000, and 10,000 yr, respectively. Since an objective of this study is to compare ^{14}C waste management options, it is worth noting that Figure 2 shows that an effective storage and containment policy would require immobilization and confinement for extremely long periods of time. For example, to reduce the eventual collective dose commitment by a factor of 10, ^{14}C waste would have to be immobilized for nearly 20,000 years. It is also evident from the results that, if the ^{14}C was immobilized and disposed of to the deep ocean, it would delay delivery of the dose but would not actually reduce it by any significant fraction. For the reactor distribution summarized in Table 1, ~97 Ci of ^{14}C are produced per GW(e) year of electricity (this value exceeds the 47 Ci/GW(e) year figure used in calculating the releases to 2050 because it includes solid wastes which may not be released to the environment until after reactor decommissioning, *ie,* after 2050). Thus, 1 GW(e) year of electricity produced by nuclear power would lead to a collective dose commitment of ca 450 man Sv from the subsequent ^{14}C discharges.

Although atmospheric CO_2 specific activities are not expected to alter

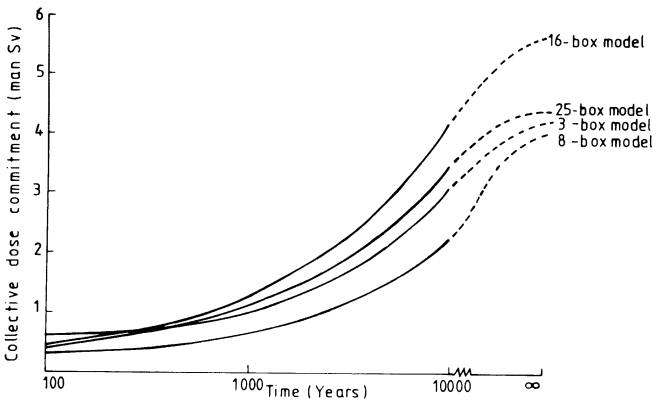

Fig 2. Development of collective dose commitment arising from the release of 1Ci of ^{14}C into the atmosphere

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significantly in the period up to 2050, the subsequent collective dose commitment to the world population is high in comparison to those from other waste nuclides. To reduce the eventual dose to the public, in-plant ^{14}C clean-up procedures, with subsequent immobilization of the solid ^{14}C waste, would be required. However, this task will be rendered difficult and expensive by the long storage and containment periods involved.

LOCAL EFFECTS OF ^{14}C DISCHARGES

It is to be expected that exposures to ^{14}C of individuals in the vicinity of large nuclear installations will exceed the average for the general population. To help quantify this effect and to assess the efficiency of atmospheric dispersion models, biologic samples have been collected for the last three years in the vicinity of British Nuclear Fuels plc nuclear fuel-reprocessing plant at Sellafield (Windscale) in Cumbria, the largest artificial source of ^{14}C in the U K. The samples (mainly grass) were collected along a northeasterly transect (that shown by Otlet, Walker & Longley (1983) to have the highest ^{14}C activity in 1981). Clearly for conservative modeling purposes, this transect is the one of major significance and our interest here was to determine whether any mathematical model could satisfactorily account for downwind concentration gradients over successive years of continuous discharge. In the absence of published data, on ^{14}C levels in vegetation, in British Nuclear Fuels plc reports, sample collection and analysis were necessary. Samples were analyzed at Glasgow by conversion to benzene and subsequent liquid scintillation counting (Campbell & Baxter, 1979). All results are corrected for isotopic fractionation and have typical associated errors of 0.7% (1 σ).

The results for 1984 are quite typical of the observed spatial distributions of excess ^{14}C. A maximum level of 27.2 $pCig^{-1}C$ (~350% above natural) was observed <1km from the release point, with enhanced activities measurable up to 29km.

In an attempt to explain the relationship between observed ^{14}C levels and distance from the plant, a Gaussian plume atmospheric dispersion model was used (Clarke, 1979). The model assumes that the vertical dispersion of activity may be described by a Gaussian distribution, while horizontal dispersion is uniform across a sector of angle α (usually $\alpha = \pi/3$ radians) for a continuous release. Assuming a constant release rate from Sellafield, the model can be used in combination with site-specific data (stack height, meteorologic data, etc) to predict the spatial distribution of the excess ^{14}C. The meteorologic data used here were the average distribution of the wind rose and the Pasquill stability categories, in the sector of interest, for the period April–September inclusive. These data were obtained from the Blackpool Met Office station ca 70km south of Sellafield.

Since the meteorologic data for 1984 are not yet available, the model predictions for 1983 are shown in Figure 3, along with the observed ^{14}C activities for that year (with the baseline value of 7.5 $pCig^{-1}C$, measured by assay of grass collected from a remote site in northwest Scotland, subtracted). The reliability of the release rate assumed in the model is difficult to assess since BNF plc are unsure of the validity of their published release

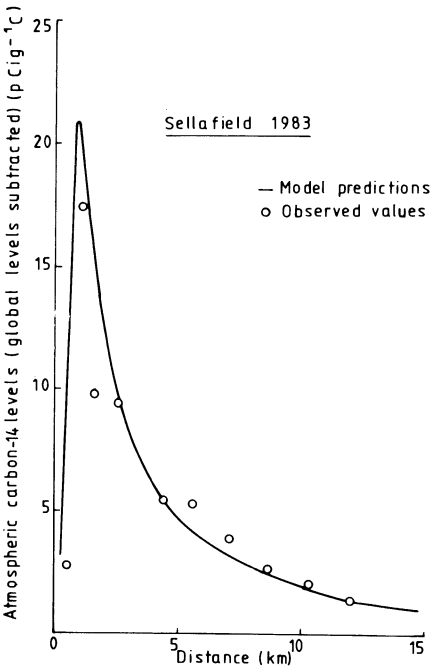

Fig 3. Excess atmospheric ^{14}C levels up to 12km from Sellafield

rates (British Nuclear Fuels plc, 1982, 1983, 1984). However, it is clear from Figure 3 that agreement between the observed and predicted values is excellent suggesting that the Gaussian plume model provides a realistic description of atmospheric dispersion.

As the distance from the discharge point increases, the governing equation of the Gaussian plume model tends towards a simple hyperbolic function. Therefore, the downward trend in the above model can be adequately described by the function

$$C = \frac{K}{R}$$

where C is the atmospheric ^{14}C level ($pCig^{-1}C$), R is the distance from the release point (km) and K is a constant which is directly proportional to the release rate. The ^{14}C levels for the years 1982–84 have been plotted in Figure 4A *vs* ^{1}/distance. Here, the ^{14}C levels have been adjusted to a normalized wind rose so that any differences in the observed ^{14}C gradients must be solely due to fluctuating release rates and not to fluctuating wind directions. The results obtained by Otlet, Walker and Longley (1983) for the same transect in 1981 have also been plotted for comparison. All lines show high degrees of linear correlation (demonstrated in Figure 4B for the 1984 results) with the ordinate intercepts clearly indicating the existence and magnitude of the global baseline levels. The intercepts for the years 1982–84 (from grass samples) were all very similar, in the range 7.7 ± 0.4

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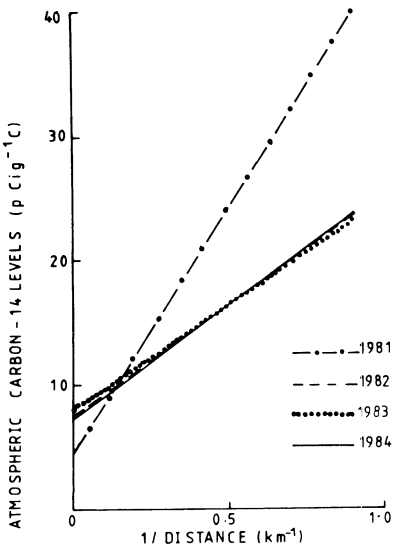

Fig 4A. Trends in atmospheric ^{14}C levels *vs* distance^{-1} for the Cumbrian environment in 1981, 1982, 1983, and 1984 (1981 data from Otlet, Walker and Longley (1983))

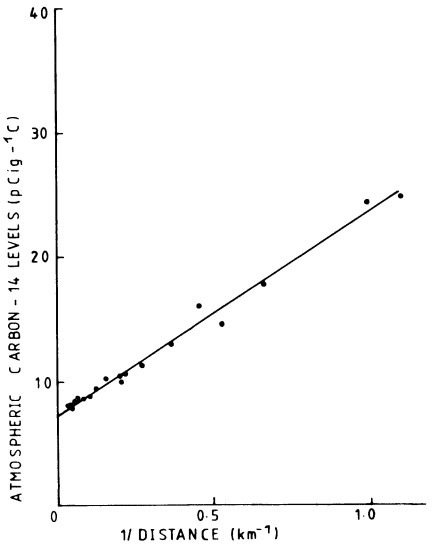

Fig 4B. Atmospheric ^{14}C levels *vs* distance^{-1} for the Cumbrian environment in 1984

pCig^{-1}C, and are very close to the actual baseline values. The 1981 value, however, (from hawthorn berry samples (Otlet, Walker & Longley, 1983)) is much lower (4.8 pCig^{-1}C). This deviation may suggest that hawthorn berries are not as precise indicators of ambient atmospheric ^{14}C levels or, more likely, it may reflect the simplistic nature of the model and the analytical uncertainty of the data. The gradient of the lines and hence, the release rate from year to year clearly differ and are in the proportion of 7:3:3:3: for the years 1981, 1982, 1983, and 1984, respectively. These results correlate well with the data of BNF plc (British Nuclear Fuels plc, 1982, 1983 and 1984; Atherton, pers commun, 1985) which suggest that the annual ^{14}C release rates from Sellafield during this period were in the proportion of 8:4:3:3. Thus, the Gaussian plume model and its derivative simplification are, perhaps surprisingly, extremely effective in predicting the down-wind distribution from this continuous release.

However, despite the significant enhancement of ^{14}C levels around Sellafield, it would appear that the radiologic impact on the local population is negligible. In the entirely unlikely case of an individual whose total dietary intake of carbon was of the highest observed activity in 1984, then the annual intake, by ingestion, of excess ^{14}C would correspond to 1.6% of the Annual Limit on Intake for members of the public (on the assumption that the ALI for the public is of the order of 10% of that for workers as defined by ICRP (1979)).

CONCLUSIONS

^{14}C discharges from Sellafield provide an excellent tracer for the study of atmospheric dispersion models. Results thus far show that the Gaussian

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plume model performs successfully. The radiologic hazard presented to populations in the vicinity of this discharge is low.

However, because of its ease of incorporation into the global carbon cycle, together with its long half-life, the release of one unit of ^{14}C leads to a large collective dose commitment relative to other waste nuclides.

Although the specific activity of atmospheric CO_2 is not expected to deviate greatly from its present level over the next 70 years, ^{14}C discharges from the nuclear fuel cycle cannot be ignored in the longer term. Collective dose commitments, for the full range of possible waste treatment and disposal options, must be quantified and combined with other (social and economic) factors to determine the most appropriate and reasonable course of action.

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