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Sea-to-land transfer of technetium-99 through the use of contaminated seaweed as an agricultural soil conditioner

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Abstract

The use of seaweed as an agricultural soil conditioner gives rise to a potential pathway for the transfer of Technetium-99 (^{99}Tc) from marine to terrestrial ecosystems and thence to man. However, to date there is little information on the extent of the release of ^{99}Tc from seaweed into soil and the mechanisms involved. This pot experiment has shown that ^{99}Tc is released fairly rapidly from *Fucus vesiculosus* into a sandy coastal soil. Despite low temperature conditions, 60% of the ^{99}Tc added with the seaweed had accumulated in the soil 15 weeks after addition. Concurrent CO_2 monitoring (used as a measure of microbial decomposition or catabolism) suggested that the initial ^{99}Tc release (up to 40% in the first 8 weeks) was due to leaching from the seaweed and that microbial decomposition was responsible for the release of the remaining ^{99}Tc in the latter phase (12–15 weeks).
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1. Introduction

Technetium-99 (^{99}Tc) is a high yield fission product. A nuclear reactor produces about 40 kg of ^{99}Tc per annum (Leiser, 1993) most of which is recovered from the spent fuel at reprocessing plants. Its main route into the marine environment is through the discharge of effluents from nuclear reprocessing plants (Schulte and Scoppa, 1987). As a long-lived radionuclide with a physical half-life of 213,000

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years, it has the potential to contaminate the marine environment for many generations.

British Nuclear Fuels Limited (BNFL) at Sellafield in Cumbria, UK has been authorised by the UK government to discharge controlled amounts of radionuclides, including ^{99}Tc, into the Irish Sea since 1952 but the commissioning of the Enhanced Actinide Removal Plant (EARP) in 1994 has led to an increase in annual discharges of ^{99}Tc (Leonard et al., 1997). EARP was built to remove alpha and beta activity from stored concentrated waste streams and other effluents, but ^{99}Tc cannot be effectively removed as it is present as the highly soluble pertechnetate ion (TcO_4^-)in the acidic effluent (Leonard et al., 1997). In 1998 it was estimated that 1260 TBq of ^{99}Tc remained in storage at Sellafield (Brown et al., 1998). Despite the annual discharge limit being cut from 200 TBq to 90 TBq in 1999 and further decreases planned for the future (EA, 2000), ^{99}Tc discharges into the Irish Sea are likely to continue for many decades unless a suitable abatement technique can be found. It has been shown that once in the marine environment ^{99}Tc is bioaccumulated by brown seaweed, particularly *Fucus* species and *Ascophyllum nodosum*, with concentration factors for *Fucus* species in excess of 1×10^5 (wet weight) (Brown et al., 1998). The level of ^{99}Tc in *Fucus vesiculosus* in the Irish Sea close to Sellafield has risen over 50-fold since the commissioning of EARP (Camplin et al., 1999).

The accumulation of ^{99}Tc by brown seaweed gives rise to a potential pathway for the transfer of ^{99}Tc to terrestrial ecosystems and on to humans, through the practice of using seaweed as an agricultural soil conditioner in coastal areas (Camplin et al., 1999). This practice, whilst not as widespread as in the past, still occurs on a small scale on the west coast of Britain, including Cumbria. A study of plots where seaweed was used in Cumbria, close to the Sellafield plant, showed that whilst ^{99}Tc activity in root vegetables (e.g. onions 8–26 Bq kg^{-1}) was low, the concentration in leafy vegetables such as spinach was high (8,400 Bq kg^{-1}) (Camplin et al., 1999).

Some parts of the sea-to-land pathway have been well researched such as the uptake of ^{99}Tc by brown seaweed (Beasley and Lorz, 1986; Holm et al., 1986; Smith et al., 1997; Brown et al., 1998) and the uptake of ^{99}Tc (in the soluble pertechnetate ion form) from soil by certain plant species (Wildung et al., 1977; Lembrechts & Desmet, 1986; Van Loon, 1986; Echevarria et al., 1997). However there has been little published research on the release from plant material, including seaweed, into soil. Two studies have shown that ^{99}Tc incorporated into grass and wheat leaves was readily extractable in water and as bioavailable to plants as pertechnetate added in solution to the soil (Dehut et al., 1989; Echevarria et al., 1997) but these studies did not investigate the mechanisms involved in the release of ^{99}Tc from the plant tissues.

This paper presents the results of a pot experiment designed to measure the release of ^{99}Tc from *F. vesiculosus* into soil under conditions similar to those encountered in the field. The specific objectives were to:

(a) quantify the amount of ^{99}Tc released from the seaweed into a sandy coastal soil over a period of 15 weeks,
(b) measure any concurrent changes in CO_2 production by soil microorganisms, and

(c) interpret the temporal release pattern for ^{99}Tc in relation to soil microbial activity and decomposition processes.

2. Materials and method

2.1. Materials

Two users of seaweed on garden plots in Cumbria were interviewed to ascertain the collection practices, application methods and crops grown so that the experimental design could mimic the actual use of seaweed as a soil conditioner.

Fucus vesiculosus (identified by the presence of pairs of vesicles on the fronds) was freshly cut from rocks on the shore close to the Sellafield reprocessing plant (Grid reference NY 018034) in January 2001. The seaweed was stored at 5 °C for 7 days prior to its addition to the soil.

Soil was collected in an area of unimproved grassland on the east coast of Scotland (Grid reference NO 470237) by removing the vegetation and sampling at a depth of 10–15 cm. The material is classed as freely draining, stabilised, wind blown sand (Soil Survey of Scotland, 1980) (pH 4.1, 2.8% organic matter) and was similar to the type of soil to which seaweed is added in Cumbria. The field moist soil was sieved through a coarse sieve to remove root material and stored at ambient temperature (<5 °C) before use.

2.2. Experimental design

A series of 1 l pots were prepared with 50 g (wet weight) *F. vesiculosus* chopped into 4–6 cm pieces, mixed evenly with 250 g (field moist) ^{99}Tc-free (confirmed by analysis) sandy coastal soil. To estimate the amount of ^{99}Tc added to each pot a 100 g sample of seaweed was divided into equal halves by cutting individual fronds in two. One half of the sample was added to the pot and the other retained for ^{99}Tc analysis. Eighteen test pots and 18 controls (soil only) were set up, kept at ambient temperature in an open-sided shelter at Stirling University (Grid reference NS 817969), and maintained at 65% field capacity (measured gravimetrically) by watering regularly with rainwater. The ambient temperature on campus was recorded at the University's weather station. The experiment ran for 15 weeks from the beginning of February to May 2001. On six occasions, three test and three control pots were removed from the experiment for sampling.

2.3. CO_2 measurements

At each of the six sampling dates CO_2 production by soil microorganisms over a 24-h period was measured. This was done by capping the pots with airtight lids, immediately taking 1 ml samples from the air space in the pots (base measurements), then taking further 1 ml samples 24 hours later. The air samples were analysed for CO_2 concentration by gas chromatography using a Varian Aerograph, model 90-P.

As CO_2 production is widely accepted as a qualitative expression of active decay processes (Wagner and Wolfe, 1998) an increase in CO_2 production was assumed to represent an increase in microbial decomposition.

2.4. ^{99}Tc analysis

On each sampling date the seaweed pieces, which were still largely intact, were removed manually from the pots and the soil dried at 60 °C. The soil was thoroughly mixed and analysed for ^{99}Tc using the method developed for environmental samples by Wigley et al. (1999). This method has a detection limit of 1.7 Bq kg^{-1} based on a 2 h count time and a 10 g sample. The same technique was used to analyse the seaweed samples retained at the start of the experiment. This involves ashing the sample, acid leaching, removal of contaminants present in environmental samples, e.g. Fe, ^{60}Co, ^{137}Cs, through the use of precipitation and anion-exchange chromatography and finally the extraction of ^{99}Tc in an organic solvent. A known activity of ^{99m}Tc (γ emitter, half-life 6.06 h) was added at the acid digestion stage of the process as a yield monitor. Tc-99m activity in each sample at the end of the process was measured on an Ortec 35% relative efficiency n-type HPGe counter and compared with a ^{99m}Tc standard to determine the recovery rate. The recovery of ^{99}Tc was assumed to be the same as that for ^{99m}Tc. The method was validated using five *Fucus* species samples provided by the Scottish Universities Research and Reactor Centre, East Kilbride, UK that had previously been used in an international intercomparison exercise (McCartney et al., 1999). In addition, an internal seaweed standard (*F. vesiculosus*) was included in every batch of samples analysed. The activity of ^{99}Tc in the samples was measured by liquid scintillation counting on a Wallac 1220 Quantulus Ultra Low Level Liquid Scintillation Counter.

2.5. Statistical analysis

Statistical analysis was carried out using MINITAB (Minitab, 2000). A two-way ANOVA was performed on square root transformed CO_2 production data, using a General Linear Model with ‘sample date’ (i.e. time) and ‘treatment’ (i.e. with or without seaweed) as factors. A one-way ANOVA was performed on square root transformed ^{99}Tc in soil (% of that added with seaweed) using ‘sampling date’ as the factor. This data was further analysed using a Tukey’s pairwise comparison (family error rate 0.05) between sampling dates.

3. Results

3.1. User interviews

The individuals interviewed collected drift seaweed from the strandline mainly in late autumn through to early spring, after stormy seas had cast the seaweed on to the beach (Anonymous, 2000). Several species are collected but the bulk of the

collection is *Fucus vesiculosus* which is a common species along that coast. The seaweed may be added directly to the soil in early spring or composted with other plant material before addition to the soil. The soil in the area has a sandy texture with little organic matter. A variety of vegetables are grown on these plots, such as potatoes, beetroot, spinach and cabbages.

3.2. CO_2 production

The difference in CO_2 concentration between the base and 24-h samples of air from the sealed pots was taken as representing the amount of CO_2 produced by soil microorganisms in 24 h. The CO_2 was calculated as the number of moles of CO_2 per gram of soil (dry weight) produced in the 24-h period. The results obtained were plotted (Fig. 1) to show the change in CO_2 production rate over time for the treatment (with seaweed) and control (without seaweed) pots. The graph also shows the ambient daily temperature over the period of the experiment.

The rate of CO_2 production in the control pots remained low and fairly constant throughout the experiment. Over the first three weekly sampling points (1–20 February) the CO_2 concentration in the treatment pots remained low and there was little difference in CO_2 production between the treatment and control pots. CO_2 production had risen 10-fold by the sampling point at the end of March (8 weeks). The rate fell substantially at the 12 week sampling point at the end of April before rising again at the final sampling point in May (15 weeks) to a level approximately equal to that found at the 8 week sampling point. Statistical analysis confirmed that there was a significant effect of time and treatment ($p<0.01$) on CO_2 production and the interaction of treatment and time was also significant ($p<0.01$). The total amount of CO_2 released over the 15-week period was calculated to be 2×10^{-2} moles.

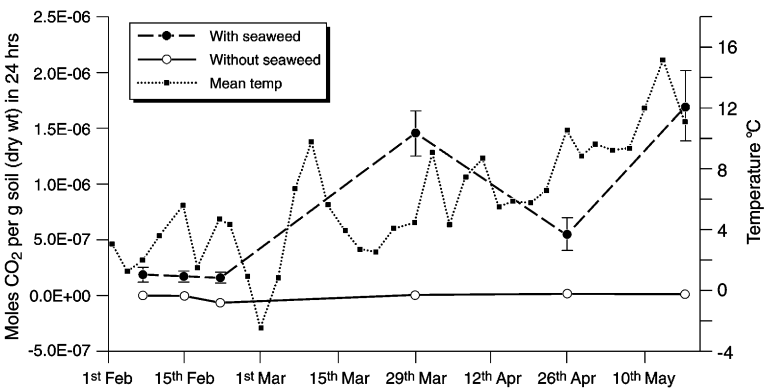

Fig. 1. Carbon dioxide production (moles per gram soil, dry weight) in treatment and control pots over time ($n = 3$).

Table 1
Results from validation exercise

Sample	Mean (Bq kg^{-1}) [a]	SD (Bq kg^{-1}) [a]	This study (Bq kg^{-1})
A	5.86	3.53	7.18
B	58.27	16.57	66.99
C	2,910	500	2,699
D	17,910	2,700	16,680
E	133,100	18,000	147,371

[a] 14 laboratories (McCartney et al., 1999).

3.3. Accuracy of ^{99}Tc analysis

The results from the analysis of the validation samples are shown in Table 1. All results from the analysis for this study fell within the one standard deviation of the mean results from other laboratories in the exercise. Analysis of the internal seaweed standard gave a mean result of 5.93×10^4 Bq kg^{-1} (dry weight) ($n = 8$, SD = 0.594×10^4).

3.4. ^{99}Tc in seaweed and soil

F. vesiculosus samples (added to the pots at the start of the experiment) had a mean activity concentration of 5.95×10^4 Bq kg^{-1} (dry weight) (1.19×10^4 Bq kg^{-1}, wet weight) ($n = 18$, SD = 0.149×10^4). The measured values can be compared with those for *F. vesiculosus* collected from the shore close to BNFL, Sellafield in previous years in Table 2.

Analyses of the soil samples collected between 5 and 105 days (15 weeks) after the start of the experiment showed that ^{99}Tc had been released from the seaweed into the soil. Changes in ^{99}Tc activity in the soil over time (assuming even

Table 2
^{99}Tc concentration in *Fucus vesiculosus* close to BNFL Sellafield, Cumbria, UK

Year	Bq kg^{-1} (wet weight)	Reference
1997	60,000	MAFF[a] & SEPA[b] (1998)
1998	20,000	MAFF & SEPA (1999)
1999	13,000	FSA[c] & SEPA (2000)
2000	10,000	FSA & SEPA (2001)
2001	11,850	STIRLING

[a] Ministry for Agriculture, Fisheries and Food
[b] Scottish Environmental Protection Agency
[c] Food Standards Agency

Table 3
^{99}Tc concentration in soil over time (n = 3)

Week of experiment	^{99}Tc Bq kg^{-1} (dry weight)	SD (1σ)
0.7	147	57
1.7	297	153
3	423	131
8	1153	421
12	932	225
15	1691	631

distribution) are given in Table 3. The underlying assumption that all seaweed had been removed from the soil is based on the observation that the frond pieces appeared intact. Very small fragments potentially not removed, would have no significant effect on the result. Very small fragments potentially not removed, would have no significant effect on the result. The ^{99}Tc released into the soil in each pot was calculated as the percentage of that originally added with the seaweed. The results are plotted as the cumulative percentage of the original ^{99}Tc added to show the pattern of accumulation over time (Fig. 2). About 20% of the ^{99}Tc added to the pot with the seaweed had accumulated in the soil over the first three weeks of the experiment. By the 8-week sampling point this had increased to around 40% and there was no further increase in this value by the 12-week sampling point. By the end of the experiment (15 weeks) over 60% of the ^{99}Tc originally added with the seaweed had been transferred to the soil. There was a statistically significant overall effect of time on ^{99}Tc levels in soil ($p<0.05$). Multiple comparisons showed no significant difference in the ^{99}Tc levels between the first three sampling dates but levels from sampling dates four and six were significantly higher than those from the first three dates.

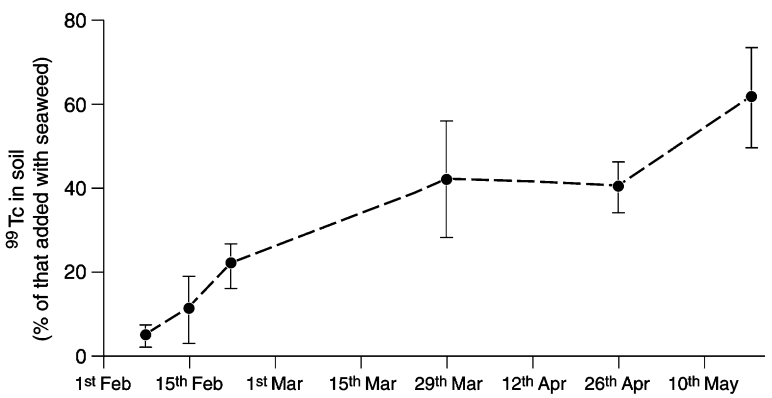

Fig. 2. ^{99}Tc accumulation in soil (cumulative percentage of that added within seaweed) over time (n = 3).

4. Discussion

This experiment attempted to mimic the use of seaweed on garden plots in Cumbria through the use of a sandy coastal soil and representative application rates and techniques for the seaweed. However, because of the small size of the pots, the application rate of the seaweed had to be reduced (by about 50%) and the seaweed cut up to allow its incorporation into the soil. The seaweed also had to be freshly cut from rocks as there was none present on the strandline at the time. The timing of the application was realistic as the growers often incorporate the seaweed early in the year before planting their vegetables in early spring.

The ^{99}Tc activity concentration in *F. vesiculosus* in the vicinity of BNFL Sellafield has fallen since 1997 (Table 2) reflecting the fall in ^{99}Tc discharges from the reprocessing plant (Brown et al., 1999). The seaweed collected for this experiment shows a slight increase in ^{99}Tc concentration compared to the 2000 value. This may be due to the different sample collection methods. In the RIFE (Radioactivity in Food and the Environment) reports, seaweed was collected at four different times throughout the year and the samples bulked for ^{99}Tc analyses as opposed to the single sample collected in winter for this experiment. ^{99}Tc concentration in seaweed has been found to be higher in the autumn and winter (Kershaw et al., 1997) so this could explain the increase in this study from the 2000 RIFE level.

The results from this experiment were analysed to determine the relative importance of three processes involved in decomposition; commutation (mechanical reduction of particle size), leaching and catabolism (energy producing, enzymatic transformation of complex organic molecules into simple ones, e.g. by soil microbes) (Swift et al., 1979), for the release of ^{99}Tc from contaminated seaweed incorporated into soil. These processes are regulated by many factors including the type and number of decomposer organisms in the soil, the resource quality and physio-chemical factors such as temperature, soil moisture and soil aeration (Swift et al., 1979). In the pot experiment, the soil was kept at 65% field capacity which was optimal for decomposition (Wagner and Wolfe, 1998) providing enough soil moisture and oxygen for microbe activity.

The cutting up of the seaweed prior to its addition to the soil mimics commutation of the resource. This may destroy cell walls allowing the escape of soluble material and give a larger surface area for microbial action. Carbon dioxide production is widely accepted as a quantitative expression of catabolic processes (microbial decomposition) (Wagner and Wolfe, 1998). In the first 3 weeks, CO_2 production in the pots containing seaweed remained low, similar to those in the control pots, suggesting that little, if any, catabolism of the seaweed was taking place, probably due to the low ambient temperature (2–6 °C) over this period. Despite this, around 20% of the ^{99}Tc added with the seaweed had accumulated in the soil over this initial period. This may be due to leaching, an abiotic process, of soluble pertechnetate ions and Tc complexed with various low molecular weight ligands within the cell, possibly enhanced due to the cutting up of the seaweed frond prior to their addition to the pots. About 50–70% of ^{99}Tc in plants is in an ionic form or associated with soluble proteins or small organic molecules (Cataldo et al., 1986).

Although the cumulative percentage of ^{99}Tc accumulated in the soil had increased to 40% by week eight the overall rate of release from the seaweed between three and eight weeks was slower than that in the first three weeks. The 10-fold increase in CO_2 production at this point does not seem to be a temperature response (as the temperature over this period generally did not exceed 6 °C), so it may represent the microbial decomposition (catabolism) of leached soluble sugars, with the slowing release rate of ^{99}Tc representing the continuing leaching of the remaining soluble forms of the radionuclide. At 12 weeks the CO_2 production rate fell considerably which may indicate the depletion of the leached, readily decomposable material (again there is no obvious response to temperature). Between eight and 12 weeks after seaweed addition no ^{99}Tc appeared to be released suggesting that the remaining ^{99}Tc was bound to cell organelles such as chloroplasts (Woodard-Blankenship et al., 1995) or large insoluble molecules that could not be leached out of the cells. At the last sampling point (15 weeks), CO_2 production had risen again. This may partly be a response to a steady increase in ambient temperature but also may indicate commencement of catabolism of less readily decomposable material, perhaps some of the insoluble storage molecules within the cells. This was accompanied by a further increase in ^{99}Tc released into the soil suggesting that some of the bound ^{99}Tc was being released due to the catabolism of large organic molecules with which they had formed complexes. By the end of the experiment over 60% of the ^{99}Tc added with the seaweed had accumulated in the soil. This was much higher than expected considering the low ambient temperature at the time, the short duration of the experiment and the intact appearance of the seaweed pieces on their removal from the pots at every sampling date. There was a lack of information on the percentage release of ^{99}Tc from seaweed or any other plant material in published literature with which to compare the results from this study.

The amount of carbon released as CO_2 over the period of the experiment equalled approximately 8% of the carbon content of the seaweed, assuming that the carbon content of *F.vesiculosus* is equal to that reported (6% of fresh weight) for *F. serratus* (Bidwell and McLachlan, 1985). This suggests that large-scale decomposition of the structural carbohydrates of the seaweed had not yet taken place.

Two-compartment kinetics for the release of ^{99}Tc into water from plant material (including brown seaweed) have been previously reported (Benco et al., 1986; Dehut et al., 1989) but to date there have been no reports of studies into the kinetics and mechanisms involved in the release of ^{99}Tc from seaweed into soil. The results of this experiment, ehich are in agreement with the previously reported two-phase release patterns, suggest that the ^{99}Tc release from seaweed into soil is the product of two mechanisms, leaching and microbial decomposition (catabolism).

5. Conclusion

Under the winter conditions existing in the initial phase of the experiment, leaching appears to be the predominant mechanism for the release of ^{99}Tc from seaweed into soil. Microbial decomposition seems to become more important as ambient tempera-

ture rises in the spring in the latter stages of the study. Over 60% of ^{99}Tc contained within the seaweed was released into the soil between the beginning of February and mid-May and a similar release can reasonably be expected under field conditions in Cumbria to coincide with the start of the growing season. It therefore seems likely that there will be a high availability of released ^{99}Tc for plant uptake under aerobic conditions.

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