EXPERIMENTAL STUDIES ON THE BEHAVIOUR OF TECHNETIUM IN THE MARINE ENVIRONMENT**

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Introduction

The presence of technetium in the biosphere depends largely on the most important isotope ^{99}Tc, which enters the environment as a result of nuclear fuel reprocessing, nuclear waste storage, nuclear weapon testing and, to a very limited extent, the use of ^{99m}Tc in radiodiagnostics (1,2).

The long-lived radionuclide ^{99}Tc ($T_{1/2} = 2.15 \times 10^5$ years) is produced in relatively high yield (6%) in both thermal and fast fission of ^{235}U, ^{238}U and ^{239}Pu. Because of its long half-life, ^{99}Tc contributes to a considerable extent in determining amounts of long-term radioactivity and hazards of high-level wastes from nuclear fuel. A critical view on the inventory of the most important radionuclides in spent fuel from light water reactors shows clearly that after nearly 1000 yeras of aging, ^{99}Tc represents the major contribution to the all-over radioactivity of waste, but in terms of hazard, ^{99}Tc is much less important even after 10^6 years because it is not as radiotoxic as the actinides.

Despite projections showing future increase in the inventory of ^{99}Tc (170,000 kg equal to 1.06×10^{16} Bq by the year 2000)(3), little information is available on the biogeochemical behaviour of technetium in the marine environment. Previous studies of the behaviour of technetium in sea water and sediments have shown (4,5) that in agreement with thermodynamic considerations, the anion pertechnetate is the chemical form most stable in well oxygenated sea water and in aerobic sediments. In the presence of anoxic sediments, pertechnetate may be reduced and immobilized as highly insoluble compounds (5). Bacterial activity and organic matter content could play an indirect role in the fixation process of technetium by determining redox conditions favourable for its immobilization in the sediments.

A review of recent studies on the behaviour of technetium in marine biota shows that ^{99}Tc is accumulated by a variety of marine organisms (6). The present investigation will give attention to the biokinetic behaviour and fate of technetium in selected marine organisms.

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Material and Methods

The photon-emitting radionuclide ^{95m}Tc ($T_{1/2}$= 60 days) facilitated greatly the experimental procedure in as much as it permitted in vivo measurements of technetium in organisms. The different gamma energies are easily detectable with NaI(Tl) scintillation crystals or by Compton effect in a liquid scintillation medium. We used both well-type crystals and an Armac (Packard Instr. Co.) liquid scintillation detector for large samples. All measurements of ^{95m}Tc were corrected for physical decay.

Experimental organisms: the crabs Pachygrapsus marmoratus, Carcinus maenas and the benthic fish Lepadogaster lepadogaster, were maintained in aquaria with running seawater for sufficiently long times to achieve good acclimatation to the experimental conditions prior to being used in radiotracer experiments.

The accumulation of ^{95m}Tc as pertechnetate (37 kBq/L) from water by the mediterranean crab P. marmoratus was followed for one month using 20 specimens (15 females and 5 males) of 12.99±4.95 g and of a carapace width of 27.85±3.86 mm. The single specimens were whole-body counted after being rinsed for several minutes in a non-radioactive bath in order to get rid of the external radioactivity of the exoskeleton and the gills. The crabs were fed daily with fresh mussels for two hours. The technetium concentration in sea water was checked at different time intervals and readjusted to the initial value by adding new pertechnetate solution.

At the end of the accumulation period, 10 specimens were dissected and the incorporated radioactivity was measured in the organs and tissues. The other 10 specimens remaining were used to follow the release kinetics of ^{95m}Tc over a period of 110 days by placing the crabs in non-radioactive water which was changed each day. Measurements of the radioactivity remaining were made by whole-body counting. At the end of the release phase, with about 10% of initial radioactivity still incorporated, the crabs were dissected and the distribution of ^{95m}Tc measured in organs and tissues.

Attempts to determine the assimilation efficiency of ingested technetium and the subsequent behaviour of its different oxidation states have been made using crabs (Carcinus maenas) and benthic fish (Lepadogaster lepadogaster). Several individuals of both species previously acclimated in running sea water were fed a single ration of mussel digestive gland which had been injected with 10 µl ^{95m}Tc solution (activities ranging from 5.5 to 18.5 kBq) in either the VII or IV oxidation states.

Immediately following ingestion, the animals were whole-body counted and replaced in glass jars containing 1 L filtered sea water which was changed daily. This procedure was continued for 4 days, after which time the crabs were transferred to large basins with running sea water. The diet of the fish was supplemented with Artemia brine shrimp. During the first 3 days of excretion, fish fecal pellets were filtered from the sea water daily and also counted for radioactivity. Crab feces were monitored in a similar fashion until the 28th day of loss.

RESULTS

Bioavailability of Technetium

The bioavailability of technetium to crabs and fish was studied in two different ways. For P.marmoratus, technetium-95m as pertechnetate was taken up from water over a one month period, while for C.maenas and for the fish species ^{95m}Tc either in VII or in IV oxidation states was administered to the animals in single rations of pretreated food.

In the case of direct uptake from water, the crabs accumulated the isotope very slowly, reaching a concentration factor of 8 after 30 days. The relative short exposure time did not permit the animals to reach equilibrium conditions.

There were at least three different pools for technetium found in the crabs: the digestive gland with 53%, the gut with 20%, and the gills, muscle and others with 27% of the total radioactivity assimilated. The absorbed fractions in these organs turned over with biological half-lives of 13, 35, and 118 days,respectively. Concentration factors for these principle organs were found to be 115, 67, 28, and 16 for hepatopancreas, stomach, gut, and gills, respectively.

When technetium was ingested by fish with food, assimilation efficiences ranged from 55-75% and the absorbed fraction of both oxidation states subsequently turned over with a biological half-life of approximately 120days. About 50% of the ingested radioactivity was absorbed across the gut by the crab C.maenas, but the turnover rate for this fraction was much more rapid than in fish, as evidenced by a biological half-life of only 60 days.

Organ and Tissue Distribution

At the end of the accumulation phase more than 70% of the total radioactivity taken up from water by the crab P.marmoratus was localized in the digestive system with hepatopancreas, stomach, and gut accounting for 53, 16, and about 2%, respectively. Most of the remaining technetium (20%) was associated with muscle tissues of legs and pincers, while only a small fraction of 3% was found in the gills,which may act as a way of entrance and elimination of technetium.

After 120 days of release the distribution pattern of technetium in the different organs of P.marmoratus changed. Still 16% of the body burden was localized in the digestive gland, while stomach, gut, gills, and muscle accounted for 14, 2,10 and 38% of the total radioactivity. In C.maenas similar figures were found after 131 days of release. In this case, digestive tract, gills and muscles accounted for 25, 8, 17 and 37%, respectively. The majority of the technetium was associated with the muscles, while by contrast the hepatopancreas and the digestive tract accumulated technetium to the greatest degree.

No significant difference of the distribution of Tc among the tissues of fish was noted between the IV and VII oxidation state. The highest concentration were found in the liver. This organ contained 25% of the body burden after 250 days of release. The heart and gall bladder also showed a marked affinity for technetium. The presence of significant amounts of incorporated technetium in all tissues after several months of release demonstrates its strong binding capacity to the tissues of vertebrates and invertebrates alike.

Excretion

During the first 3 days of loss about 18% of the technetium eliminated by the fish Lepadogaster lepadogaster was associated with the feces regardless the oxidation states (IV and VII). Most of this fraction was excreted during 24-48 hours post ingestion. The amount lost due to defecation is a minimum estimate since sequential leaching experiments with fish fecal pellets showed that as much as 80% of the technetium content could be lost within 4 hours. Nevertheless, it appears that most of the technetium initially ingested by these fish is excreted in soluble form either as urine or directly across the gills. In this case the two oxidation states of technetium exhibited a different elimination behaviour. About 40% of the reduced form (IV) and about 50% of the pertechnetate (VII) were excreted as soluble forms. Despite the high fecal excretion of technetium over the first few days of depuration, some residual radioactivity was found in feces excreted as long as 21 days after ingestion.

With the crabs C.maenas, the fraction lost via fecal excretion appears to be much less (about 8%) than that from fish. Furthermore, unlike fish fecal excretion of technetium occurs over a longer period as evidenced by significant amounts of ^{95m}Tc measured in crab-fecal pellets collected periodically during the first month of loss. Given the low fraction (about 8%) lost with the feces over the 28 day period, it seems clear that defecation by crabs is a less important route of technetium excretion than in fish, at least during the early phases of radionuclide depuration.

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