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Biomagnification of radiocesium in a marine piscivorous fish

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ABSTRACT: Radiocesium is the only trace element apart from Hg that may be potentially biomagnified at the top of the marine planktonic food chain. We quantified the assimilation efficiency from ingested prey, uptake rate from the aqueous phase, and efflux rate of radiocesium in a marine piscivorus fish (the mangrove snapper *Lutjanus argentimaculatus*). Aqueous ^{137}Cs exhibited an approximately linear uptake pattern over a 4 d exposure period, and was immediately transported to the muscles. The calculated uptake rate constant (0.00145 l g^{-1} d^{-1}) was independent of the ambient Cs concentration. Salinity variation appeared to have no influence on the ^{137}Cs influx within the range of 20 to 30 psu, but the influx rate increased when the salinity was further reduced to 15 psu. The assimilation efficiency in fish ingesting different prey (copepods, *Artemia*, clam tissues, and herbivorous fish), measured by a pulse-chase feeding technique, ranged between 78 and 95%. The efflux rate constant of ^{137}Cs in fishes following uptake from the dissolved and dietary phases ranged between 0.020 and 0.023 d^{-1}. The higher efflux rate in marine fishes compared to those in freshwater fishes may have been due to the ionic regulation in marine teleosts (e.g., high excretion rate to counteract the high ambient K^+ concentration). Using a simple kinetic model, we show that the dietary uptake of ^{137}Cs plays a dominant role when the concentration factors of ^{137}Cs in prey range between 50 and 100. At a lower value for the concentration factor (10), ^{137}Cs bioaccumulation in fish is dominated by uptake from the aqueous phase. The predicted trophic transfer factor (concentration in the predator to concentration in the prey) in the predatory fish ranges between 1 and 4.4 (with a median value of 2), and is consistent with the field measurements of trophic transfer factor of ^{137}Cs in the piscivorous fishes in both marine and freshwater systems. Thus, the biomagnification of ^{137}Cs in marine predatory fishes is largely caused by the extremely high ^{137}Cs assimilation from ingested prey, despite the relatively high efflux rate of ^{137}Cs compared to those measured in freshwater fishes.

KEY WORDS: Radiocesium · Biomagnification · Fish · Trophic transfer · Exposure

INTRODUCTION

Radiocesium (such as ^{134}Cs and ^{137}Cs) is discharged into the aquatic environments by nuclear facilities as a fission product (Park et al. 1983, Mount et al. 1994). The Chernobyl nuclear reactor accident raised substantial interest in the environmental fate of radiocesium due to its persistence, e.g., long half-life, low partitioning in sediments, and thus a long residence time in the water column. There are also substantial concerns for radiocesium transport in marine food chains, primarily due to its potential biomagnification (e.g., increasing concentration with increasing trophic level) at the top trophic levels, including fishes and seabirds. The concentration factor of radiocesium is generally very low at the bottom of marine food chains, e.g., 10 to 100 l kg^{-1} in marine phytoplankton (Wang et al. 2000), indicating that radiocesium is rather biologically inert at these trophic levels. Biomagnification of radiocesium at the top of the food chain has however been documented in many field observations

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(Kolehmainen 1972, Thomann 1981, Forseth et al. 1991, Rowan & Rasmussen 1993). Fisher et al. (1999) summarized the bioconcentration factor of radiocesium (concentration in the organisms to concentration in the ambient water under equilibrium conditions) in many groups of marine organisms collected from the Arctic oceans. There was a general trend for the bioconcentration factor to increase with increasing trophic level, although there were considerable variabilities within each taxonomic group.

Information on the route of radiocesium exposure in aquatic organisms is required to realistically model radiocesium bioaccumulation (e.g., accumulation from both the aqueous and dietary phases) and transport in the food chain. Kinetic modeling has recently been successfully employed in predicting the exposure pathways and concentrations of metal contaminants in marine invertebrates (Wang et al. 1996, Reinfelder et al. 1998, Wang & Fisher 1999a). There are, however, only a few reports addressing the application of kinetic modeling to trace element accumulation in marine fish, although such a model has been developed for fish by Thomann (1981). Mechanistic understanding of trace element bioaccumulation may be possible through measurements of several physiological parameters described in the kinetic model, including the contaminant assimilation efficiency (AE) from the ingested food source, the uptake rate constant from the aqueous source, and the efflux rate constant. Few measurements of these physiological parameters are available for marine fishes (Reinfelder & Fisher 1994, Ni et al. 2000).

In a recent study, we demonstrated that biomagnification in a marine benthic food chain may be marginally possible (Wang et al. 2000). For example, the trophic transfer factor (TTF) was predicted to increase with increasing trophic level, and may approach 1 in a marine benthic gastropod. The kinetic model provides a simple approach to evaluate the potential TTF, although there are considerable variations of physiological parameters described in the model under natural conditions. An understanding of the variability of these parameters is required for robust modeling of contaminant accumulation in aquatic organisms. Many field measurements on radiocesium bioaccumulation have been carried out in freshwater fishes (Garnier-Laplace et al. 2000), but there are fewer studies on marine fishes (e.g., Pentreath 1977, Kasamatsu & Ishikawa 1997). It has been shown that radiocesium can be efficiently assimilated and slowly excreted by freshwater fishes (Kolehmainen 1972, Forseth et al. 1992, Ugedal et al. 1992). Kasamatsu & Ishikawa (1997) analyzed >30 species of crustaceans, cephalopods and teleost fishes, and found that ^{137}Cs concentration increased with increasing trophic levels. The biomagnification factor (^{137}Cs concentration in predator to ^{137}Cs concentration in prey) was found to be 2.0 (1.8 to 2.2). The processes responsible for the high TTF of radiocesium in the top predatory fish remain speculative. Although earlier experimental studies investigated the uptake and depuration of radiocesium in several marine fishes (reviewed by Pentreath 1977), kinetic parameters such as the radiocesium uptake rate constant and the AE have not yet been well quantified.

In this study, we examined the bioaccumulation of ^{137}Cs in a marine predatory fish (the mangrove snapper *Lutjanus argentimaculatus*) from both the aqueous and dietary phases. The mangrove snapper is an important carnivorous fish widely cultured in subtropical and tropical waters. With the opening of the Daya Bay nuclear power station in Guangdong Province in southern China in the early 1990s, there is increasing concern for the fate of nuclear fission products released into the nearby coastal waters, including Hong Kong. Although there are extensive field studies on the ^{137}Cs radioactivity in different trophic levels and on the factors controlling ^{137}Cs bioaccumulation, few studies have delineated the exposure pathways of ^{137}Cs in the top marine fishes (Jefferies & Hewett 1971). Furthermore, most of these studies were carried out in the temperate and polar regions, while studies on species from tropical and subtropical waters are scarce. Bioconcentration (e.g., uptake from the aqueous phase) of radionuclides in fish from tropical/subtropical regions may have been different from that in their counterparts from temperate regions (Twining et al. 1996). In this study, we measured a few kinetic parameters (AE, aqueous uptake rate, and efflux rate) in the fish. A simple kinetic model (Thomann 1981) was then constructed to predict the TTF of ^{137}Cs and the exposure pathway of radiocesium in *L. argentimaculatus* under different environmental conditions.

MATERIALS AND METHODS

Juvenile mangrove snappers *Lutjanus argentimaculatus* (3.7 to 5.2 cm) were purchased from a fish hatchery in Taiwan. The fishes were maintained in an aerated seawater (25°C, 30 psu, 60 l) tank for about 1 wk prior to use in an experiment. They were fed fresh clam and/or frozen shrimp twice a day.

Cs uptake from the dissolved phase over time. Radioisotope ^{137}Cs (as CsCl, in 0.1 N HCl) was obtained from New England Nuclear, Boston, USA. Radioactive additions were 24.6 kBq l^{-1} (corresponding to a Cs concentration of 0.15 nM). Radioisotope and stable Cs (6 nM) were equilibrated for 12 h before the uptake measurements. Twenty-one fish were placed in 15 l of 0.22 µm filtered seawater containing both the

radiotracer (^{137}Cs) and stable Cs (as CsCl). At specific time intervals (8, 16, 24, 36, 48, 72 and 96 h), 3 fish were removed from the radioactive medium and rinsed by sequentially placing them in 3 seawater baths: (1) glass fiber filtered (GF/C) non-radioactive seawater for 5 min; (2) 0.22 µm filtered non-radioactive seawater containing 10 mM stable CsCl for 5 min; and (3) GF/C non-radioactive seawater for 1 min. The fish were then dabbed dry using tissue paper and their radioactivity was counted non-destructively by a NaI gamma detector at 665 keV (Wallac 1480). The radioactivity in the seawater was measured at the beginning and end of the Cs uptake period. There was no significant decrease in the radiocesium radioactivity in the water during the exposure period. After the radioactivity measurements, the fish were dissected into 3 compartments: gill, viscera, and other tissues (the remaining tissues including the bones). The radioactivity of each body part was measured. Finally, the dry weight of each body part was measured after drying at 80°C for >1 d. The concentration factor (CF′, l kg^{-1}) of radiocesium was calculated as the radioactivity in the fish to the radioactivity in the water and can be considered as a kinetic parameter (in contrast to the bioconcentration factor, which measures the radioactivity in the fish to radioactivity in the water under equilibrium conditions):

$$CF' = \frac{^{137}\text{Cs radioactivity in fish (cpm kg}^{-1})}{^{137}\text{Cs radioactivity in water (cpm l}^{-1})} \quad (1)$$

Cs uptake at different ambient concentrations. The uptake of Cs was determined at 4 different ambient Cs concentrations: 6, 30, 120 and 600 nM (added as stable Cs, CsCl). The lowest concentration was about 3× higher than the typical Cs background concentration (2.2 nM) in coastal waters (Bruland 1983). Radioactivity addition was 28 kBq l^{-1}, corresponding to a concentration of 0.18 nM. Six individual fish were exposed in 4 l of 0.22 µm filtered seawater at each concentration. At time intervals (8, 16, 24, 36 and 48 h), the fish were removed from the radioactive medium, rinsed twice (transferred from one beaker to another) with filtered non-radioactive water and their radioactivity counted non-destructively by a NaI gamma detector. Following the radioactivity measurements, the fish were returned to the radioactive medium. The radioactivity in the water was measured at the beginning of exposure and during the measurements of radioactivity in fish at each time point. At the end of the experiments, the fish were dissected into gill, viscera and other remaining tissues, and the radioactivity of each body part was measured. The dry weight of each body part was measured by drying at 80°C for >1 d.

Cs uptake at different salinities. Fish (originally from 30 psu seawater) were acclimated to different salinities (15, 20, 25 and 30 psu) over a period of 15 d prior to the kinetic measurements. In the low-salinity treatments, the fish were sequentially acclimated to the target salinity over a period of 7 d. During the acclimation period, they were fed fresh clam meat twice a day. The seawater was renewed every 2 d. The low-salinity seawater was prepared by diluting the seawater with Nanopure distilled water and filtered through the 0.22 µm membrane. Radioactivity additions were 28 kBq l^{-1}, corresponding to a concentration of 0.18 nM. Cs uptake was measured at a stable Cs concentration of 6 nM. Six individual fish were placed in 4 l of seawater containing the radiotracer and stable Cs. At time intervals (8, 17, 24, 36 and 48 h), the fish were removed from the radioactive medium, rinsed twice (transferred from one beaker to another) with filtered non-radioactive seawater and their radioactivity was counted non-destructively. Fish were then returned to the radioactive medium. Radioactivity in the water was also measured at the beginning of exposure and during the measurement of radioactivity in fish. By the end of exposure, fish were dissected into gill, viscera and remaining tissues (including bones), and the radioactivity was measured. The tissue was finally dried at 80°C for >1 d before the dry weight measurement.

Measurements of radiocesium assimilation efficiency (AE). Four diets were used: brine shrimp *Artemia* sp., copepod *Acartia spinicauda*, clam *Ruditapes philippinarum*, and silverside fish *Atherion elymus*. The *Artemia* sp. and copepods were radiolabeled with ^{137}Cs in artificial seawater containing 320 mM NaCl, 22.5 mM Na_2SO_4, 8.0 mM $CaCl_2$, 1.87 mM $NaHCO_3$, 42.4 mM $MgCl_2$ and 0.34 mM H_3BO_3 (Blust et al. 1992). *Artemia* were hatched and placed in artificial seawater containing ^{137}Cs (620 kBq l^{-1}) and a low concentration of K^+ (73 nM) for 36 h. The copepods were collected from Port Shelter and placed in artificial seawater containing ^{137}Cs (620 kBq l^{-1}) and a low concentration of K^+ (2.9 mM) for 30 h. Lower concentration of K^+ was used to enhance the radiolabeling efficiency. The *Artemia* sp. and copepods were then collected using a mesh and rinsed with GF/C seawater before being fed to the fishes. The clams were purchased from a market, placed in natural seawater containing radiocesium (148 kBq l^{-1}) for 2 d, and afterwards dissected. Only the foot and mantles were fed to the fish. The silversides, about 3.5 cm in body length, were caught from Port Shelter, Hong Kong, and placed in natural seawater containing ^{137}Cs (122 kBq l^{-1}) for 40 h before being used in the experiment.

No radioactive feces was produced by the snappers during the 40 and 90 min radioactive feeding. The radioactivity of the fish was immediately measured for each individual after the feeding. The fish were subse-

quently placed in non-radioactive water and allowed to depurate of the ingested food materials for 72 h. During this time, they were fed clams and the tank water was changed every 12 h.

Radiocesium depuration. Fishes were exposed to ^{137}Cs both in the aqueous phase and in the dietary phase. In the aqueous exposure treatment, 8 individual fish were exposed to ^{137}Cs in the aqueous phase for 5 d, as described previously. In the food exposure treatment, 16 individual fish were placed in 15 l of seawater and fed with both radiolabeled and nonradioactive clam tissues. The water was renewed each day. The fish were fed under these conditions for 10 d, then placed in non-radioactive water for 12 h. The surface-sorbed ^{137}Cs was removed by placing the fish in natural seawater for 5 min and then into 10 mM CsCl for 5 min. The radioactivity was immediately measured for each individual fish. Two individuals were dissected to determine the distribution of ^{137}Cs in gill, viscera and remaining tissue. Radioactivity was measured and the tissues dried at 80°C for 24 h. The remaining fish were then divided into 3 (following aqueous exposure) or 4 groups (following dietary exposure) and depurated in natural seawater (15 l) for 31 d. The seawater was renewed and fish were fed with clam meat daily. The radioactivity in individual fish was measured at frequent time intervals. By the end of the depuration period, all fish were dissected and the radioactivity in gills, viscera and the remaining tissues was determined. The tissues were then dried at 80°C for 1 d before dry weight measurements.

Modeling radiocesium bioaccumulation. Cs accumulation in the fish can be described by the following equation (Thomann 1981):

$$dC/dt = k_u \times C_w + AE \times IR \times C_f - k_e \times C \quad (2)$$

where C is the Cs concentration in fish at time t, k_u is the Cs uptake rate constant from the aqueous phase, C_w is the Cs concentration in the dissolved phase, AE is the Cs assimilation efficiency, IR is the fish feeding rate, C_f is the Cs concentration in ingested prey, and k_e is the Cs efflux rate constant. Under steady-state conditions, Cs concentration (C_{ss}) can be calculated as follows:

$$C_{ss} = (k_u \times C_w + AE \times IR \times C_f)/k_e \quad (3)$$

The relative importance of dissolved versus food uptake can be calculated as (Wang & Fisher 1999a):

$$R = (k_u)/(k_u + AE \times IR \times BCF) \quad (4)$$

where, R is the fraction of Cs uptake from the dissolved phase and BCF is the Cs bioconcentration factor in the prey organisms.

The TTF can therefore be calculated as (Reinfelder et al. 1998, Wang & Fisher 1999a):

$$TTF = (AE \times IR)/k_e \quad (5)$$

Thus a TTF > 1 indicates a possibility of biomagnification, whereas a TTF < 1 indicates a possibility of biominification.

RESULTS

Uptake from the aqueous phase

^{137}Cs exhibited a linear pattern of uptake within the 4 d exposure period, during which no steady state or equilibrium was reached (Fig. 1). The calculated CF′ was only 7.6 for the whole fish body after 4 d exposure. CF′ was higher for the gills and viscera, whereas the remaining tissues (mainly muscle and bones) had the lowest CF′. The y-intercepts of the linear regression between CF′ and time of exposure were not significantly different from zero, implying that the surface adsorption of ^{137}Cs was probably minimal in these fish. However, most of the ^{137}Cs (63 to 72%) was distributed in the remaining tissues, and only <10% of the ^{137}Cs was associated with the gills throughout the 4 d expo-

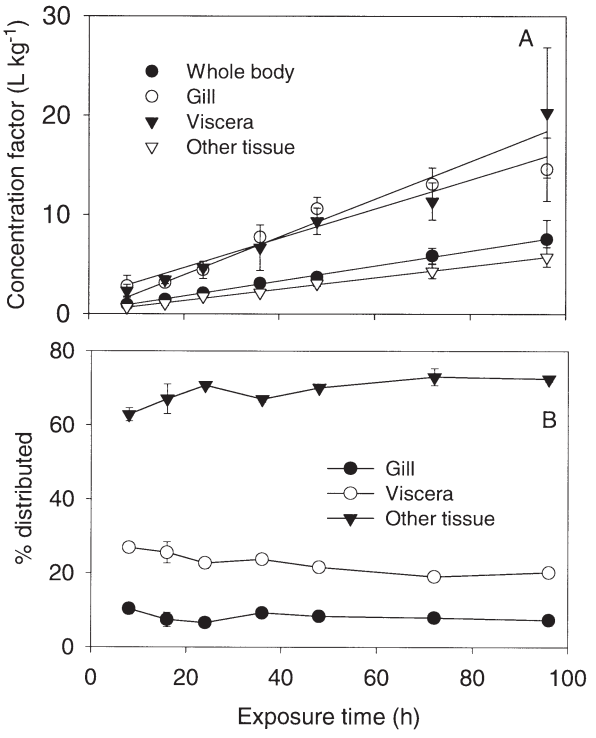

Fig. 1. *Lutjanus argentimaculatus*. (A) Calculated concentration factor and (B) distribution of radiocesium in mangrove snappers during the 4 d exposure to ^{137}Cs in the dissolved phase. Values are mean ± SD (n = 3)

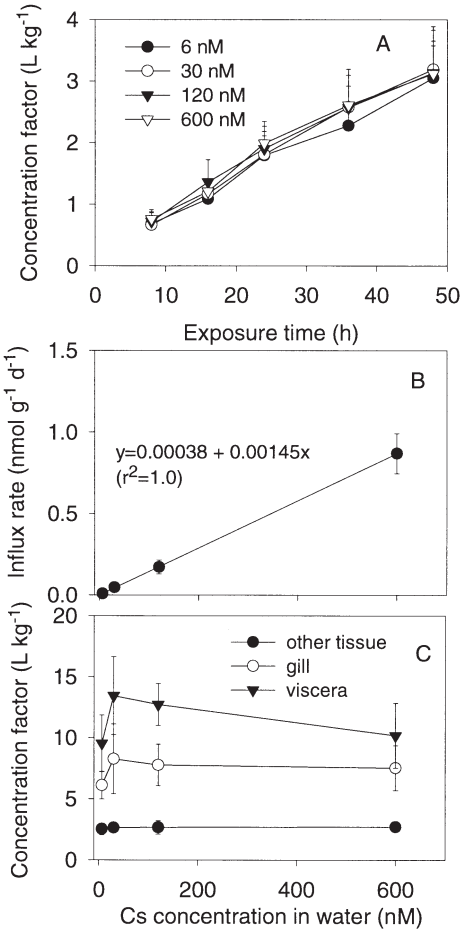

Fig. 2. *Lutjanus argentimaculatus.* Cesium uptake in mangrove snappers at different ambient dissolved Cs concentrations. (A) Calculated concentration factors at different Cs concentrations over exposure time (mean + SD; n = 6); (B) calculated influx rate as a function of Cs concentration in the dissolved phase; and (C) calculated concentration factor in different tissues of snappers as a function of Cs concentration in the dissolved phase following 8 h exposure. (mean ± SD; n = 6)

sure period. The largest amount of ^{137}Cs being in the remaining tissue may have been due to the fact that it represented the largest fraction of fish body (>80%). Further, amount of ^{137}Cs in the remaining tissue increased with increasing duration of exposure, whereas it decreased for the viscera.

Because there was a linear pattern of ^{137}Cs uptake over the 4 d exposure period, the influx rate of ^{137}Cs was measured after the exposure of fish to ^{137}Cs for 2 d. The CF′ measured at different ambient Cs concentrations (6 to 600 nM) increased linearly with time (Fig. 2). There was no major difference in the calculated CF′ at different ambient Cs concentrations. The calculated influx rate increased linearly with increasing ambient Cs concentration (with a regression coefficient of 1.0, Fig. 2). The intercept of this regression was close to zero (0.00038). Thus the uptake rate constant, calculated by the influx rate divided by the ambient Cs concentration, was 0.00145 l g^{-1} d^{-1}, and was independent of the ambient Cs concentration. In this experiment, the highest CF′ was found in the viscera, whereas the lowest CF′ was found in the remaining tissue (Fig. 2).

Salinity appeared to have no major influence on the CF′ of ^{137}Cs in the fish within the salinity range between 20 and 30 psu (Fig. 3). When the salinity was further decreased down to 15 psu, the CF′ increased substantially, although there was a large variation among different experimental individuals. For example, the influx rate increased by about 2.1 to 2.3× when the salinity decreased from 20–30 to 15 psu (Fig. 3). An ANOVA test indicated that the change in salinity did

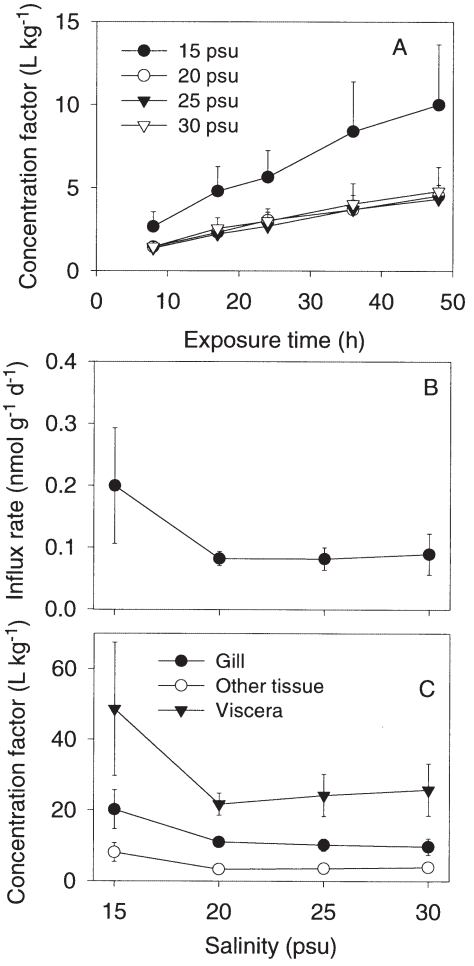

Fig. 3. *Lutjanus argentimaculatus.* Radiocesium uptake in mangrove snappers at different salinities. (A) Calculated concentration factors at different salinities over exposure time (mean + SD; n = 6); (B) calculated influx rate as a function of ambient salinity; and (C) calculated concentration factor in different tissues of snappers as a function of ambient salinity following 8 h exposure (mean ± SD; n = 6)

not significantly influence the influx rate of ^{137}Cs in the fish ($p > 0.05$). In this experiment, the highest CF′ was also found in the viscera, whereas the remaining tissue had the lowest CF′ (Fig. 3).

Assimilation of ^{137}Cs from ingested prey

The retention of ^{137}Cs in the fish following a pulse ingestion of radiolabeled food (copepod *Acartia spinicauda*, *Artemia* sp., clam *Ruditapes philippinarum*, and silverside fish *Atherion elymus*) is shown in Fig. 4.

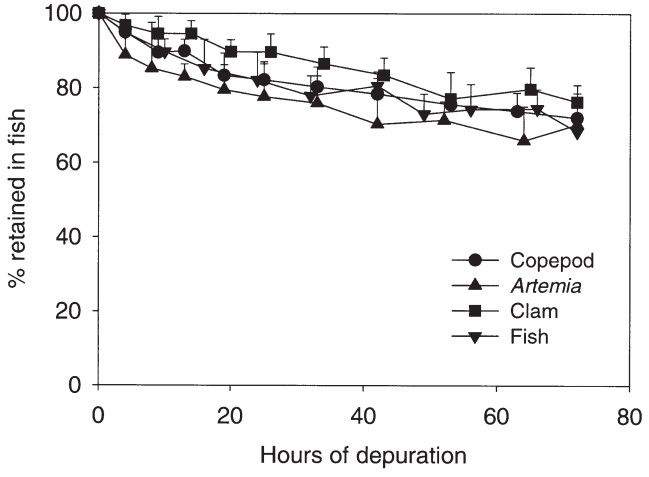

Fig. 4. *Lutjanus argentimaculatus*. Retention of radiocesium in the mangrove snappers following a pulse ingestion of radio-labeled copepods *Acartia spinicauda*, *Artemia* sp., clam tissues *Ruditapes philippinarum*, or planktivorous silversides *Atherion elymus*. Mean + SD (n = 6)

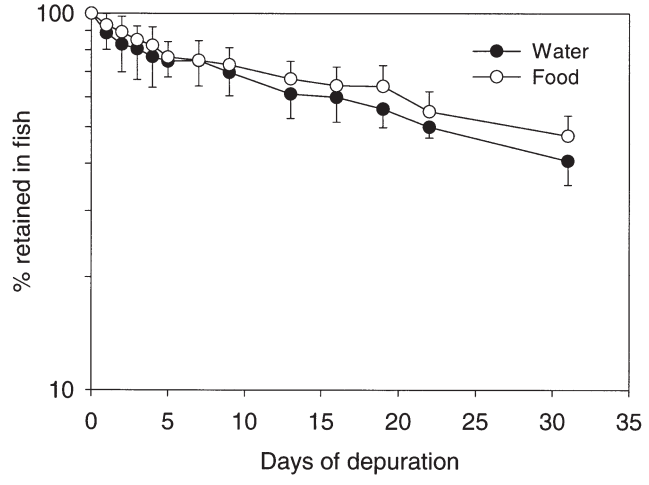

Fig. 5. *Lutjanus argentimaculatus*. Retention of radiocesium in the mangrove snappers following 5 d exposure to radiocesium in the dissolved phase or 10 d ingestion of radiolabeled clam tissues. Mean ± SD (n = 6 to 14)

Because we were not able to recover the feces for each individual fish, the time required for a complete digestion and assimilation of ^{137}Cs cannot be concluded from this study. We used the *y*-intercept method described in Wang & Fisher (1999b) to calculate the AE of ^{137}Cs in the fish, with the assumption that the loss between 24 and 72 h of depuration represented the physiological turnover of ^{137}Cs in the fish (i.e., post-digestion and assimilation). A previous study on metal uptake in marine fish generally indicated that complete digestion and assimilation required about 1 d of depuration (Ni et al. 2000). Thus, the calculated AEs of ^{137}Cs in the fish were about 88.2 ± 2.4% (1 SD) on the copepod, 77.8 ± 11.9% on *Artemia* sp., 95.1 ± 4.3% on the clam, and 89.6 ± 7.4% on the silverside fish.

Elimination of ^{137}Cs from fish

The depuration of ^{137}Cs in fish following 10 d ingestion of radiolabeled clams or 5 d exposure to ^{137}Cs in the dissolved phase is shown in Fig. 5. Loss of ^{137}Cs can be approximately described by a 2-compartmental loss (0 to 3 d and 3 to 31 d). Thus, the efflux rate constant, calculated from the slope of the ln% of ^{137}Cs in the fish against time of exposure (between 3 and 31 d), was 0.0198 ± 0.0037 and 0.0233 ± 0.0056 d^{-1} (1 SD) in fish exposed to radiolabeled food and ^{137}Cs in the dissolved phase, respectively. The calculated biological retention half-life of the slower compartment was 36.2 ± 7.5 and 31.9 ± 8.2 d (1 SD) following the exposure to food and water, respectively. There was no statistically significant difference in the efflux of radio-cesium following different routes of exposure. By the end of exposure to radiocesium, 72 to 79% of the ^{137}Cs was distributed in the remaining tissue, and 17 to 20% was distributed in the viscera (Fig. 6). After 31 d depuration in non-radioactive water, the majority of the ^{137}Cs was associated with the remaining tissues (92 to 94%), whereas only a small proportion was associated with the gills and viscera.

Modeling the exposure pathways and biomagnification of ^{137}Cs in fish

To separate the exposure route of ^{137}Cs in the fish, a few parameters are needed (Eq. 4), including k_u, AE, IR, and BCF. Values of k_u and AE are directly taken from this study. Because the AEs from different prey did not vary considerably for ^{137}Cs, we used a mean AE value of 88% (the mean of all food types considered) in our calculation. k_u was also rather constant under diverse environmental conditions (e.g., different ambient Cs concentrations and salinity). The IR of mangrove

Table 1. Numeric values of physiological and geochemical factors used in modeling the radiocesium transfer in the mangrove snappers

	Range	Mean	Source
Assimilation efficiency	0.78–0.95	0.88	This study
Ingestion rate (g g^{-1} d^{-1})	0.02–0.10	0.05	Tanasichuk et al. (1991), Jobling (1994)
Cs bioconcentration factor in prey (l kg^{-1})	50–150	100	Fisher et al. (1999)
Uptake rate constant (l g^{-1} d^{-1})	–	0.00145	This study
Efflux rate constant (d^{-1})	–	0.022	This study

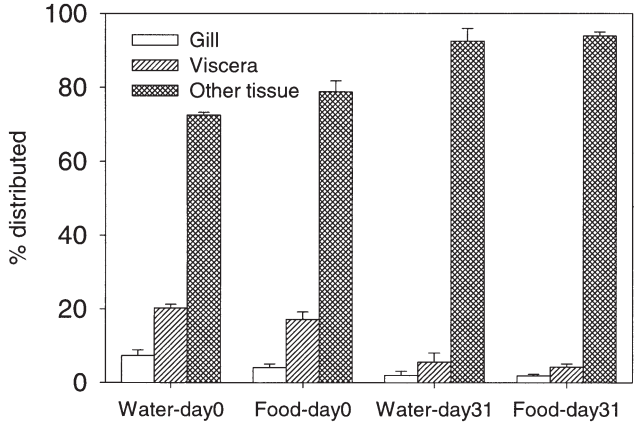

Fig. 6. *Lutjanus argentimaculatus.* Distribution of radiocesium in the mangrove snappers following 5 d exposure to radiocesium in the dissolved phase (Water-day0) or 10 d ingestion of radiolabeled clam tissues (Food-day0), and following 31 d depuration in non-radioactive waters (Water-day31, and Food-day31). Mean + SD (n = 2 for Day 0, and 6 to 14 for Day 31)

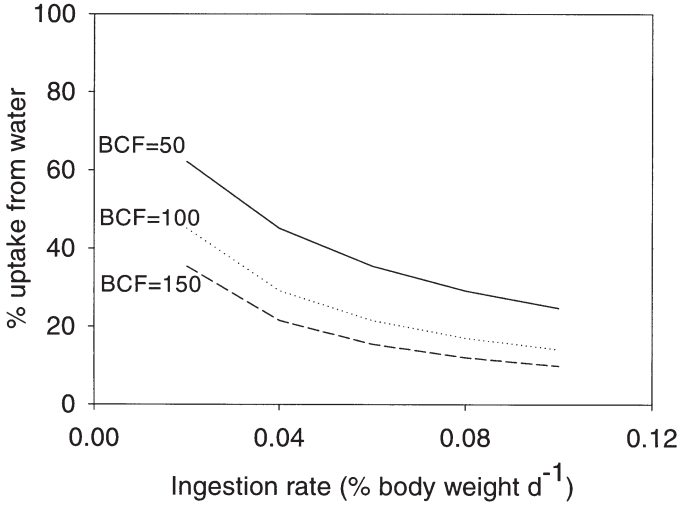

Fig. 7. *Lutjanus argentimaculatus.* Model-predicted percentage of radiocesium uptake from the dissolved phase as a function of ingestion rate of mangrove snappers. BCF: concentration factor of radiocesium in prey organisms (l kg^{-1})

snappers has not been directly quantified in the literature. The IRs (2 to 10% body wt d^{-1}, with a medium of 5%) measured in a number of carnivorous fish are used in our modeling calculations (Tanasichuk et al. 1991, Jobling 1994, Garnier-Laplace et al. 2000). The BCF, or the bioconcentration factor, in molluscs, crustaceans, and fish muscle, is taken from Fisher et al. (1999). The mean numeric value of each parameter used in the modeling is summarized in Table 1. We only performed sensitivity analysis on the influence of BCF and IR on the route of exposure in the fish. We considered 3 possible scenarios for the 2 parameters: the low-end, medium and high-end levels. Other parameters were assumed to be constant in the sensitivity analysis. Our modeling calculation indicates that uptake from the aqueous phase decreases with an increase in fish feeding rate (Fig. 7). The relative importance of aqueous versus food uptake appears to be greatly dependent on the BCF of ^{137}Cs in the prey organisms (such as zooplankton and other planktivorous fishes). At the low end of BCF for fish prey (50 l kg^{-1}), <62% of the ^{137}Cs in the predatory fish may have been derived from the aqueous uptake, whereas at the high end of the BCF (150 l kg^{-1}), <35% of the ^{137}Cs in the predatory fish may have been derived from the aqueous uptake, indicating that dietary intake of ^{137}Cs indeed dominates the total ^{137}Cs uptake in the fish. Furthermore, the calculated TTF is >1 within the normal range of fish feeding rate. The TTF is about 2 when the feeding rate is at the median level. The TTF is not related to the BCF of ^{137}Cs at the lower trophic level. Thus, our calculation indicates that ^{137}Cs may be biomagnified in top marine fishes.

DISCUSSION

The uptake of radiocesium exhibited a linear pattern over exposure time, and there was minimal surface sorption onto the fish body surface. Gill uptake may be the primary site for aqueous uptake in the fish. The radionuclide may then be rapidly transported to other tissues, as indicated by the high percentage of its distribution in viscera and the remaining tissues (mainly

muscle and bone). Similarly, radiocesium uptake in the rainbow trout *Oncorhynchus mykiss* was also approximately linear over a longer exposure period (42 d) (Baudin et al. 2000). Several lines of evidence indicate that the uptake of aqueous ^{137}Cs may be passive without active regulation. The measured influx rate of Cs into the fish bodies was directly proportional to the ambient Cs concentration. No saturation of uptake was reached within the experimental Cs concentrations tested. Thus, radiocesium may not be actively transported through the K^+ channel due to the high K^+ concentration in seawater (mM levels). The uptake rate constant (0.00145 l g^{-1} d^{-1}) in mangrove snappers was much lower than those measured in marine mussels (0.026 l g^{-1} d^{-1}) (Ke et al. 2000), presumably because of the lower ventilation (pumping) rate in the fish. Furthermore, the uptake rate was also substantially lower than those measured in the freshwater fishes (e.g., *O. mykiss*) (Garnier-Laplace et al. 2000).

The concentration factor measured in our study was generally low for mangrove snapper, consistent with many studies in other fishes or in other groups of organisms (Fisher et al. 1999). It should be noted that the concentration factor measured in marine fishes collected from the field may not necessarily reflect the bioconcentration from the aqueous phase, because radiocesium may additionally be accumulated through dietary exposure (e.g., bioaccumulation). The dominance of ^{137}Cs in the muscle tissues suggested that radiocontamination of fish tissues can potentially be a health problem, despite the low concentration factor measured for the tissues.

There are many studies on the influence of environmental factors on radiocesium bioaccumulation in freshwater fishes. Rowan & Rasmussen (1993) summarized the bioconcentration of ^{137}Cs in fishes from different freshwater systems and concluded that ^{137}Cs bioaccumulation is negatively related to ambient K^+ concentration and suspended solid loads, but is positively related to temperature. Our study indicated that salinity did not significantly affect the accumulation of ^{137}Cs in snappers until the salinity was reduced to the lower limit of physiological tolerance (15 psu). Presumably, the K^+ concentration (mM level) at these salinities was too high to cause a large change in ^{137}Cs influx with varying salinity. Acclimation at different salinities had no effect on the accumulation of ^{137}Cs by the blenny *Blennius* sp., but the uptake was related to the body size and temperature (Lucu & Jelislavčič, cited in Pentreath 1977).

Our study demonstrated that the AE of radiocesium in the predatory fishes was exceedingly high (78 to 96%). To our knowledge, this is the highest AE reported for ^{137}Cs in fish. For example, Hewett & Jeffries (1978) found that the AE of ^{137}Cs from food was 42% in plaice and 67% in trout. Recently, Cocchio et al. (1995) measured an AE of 65% in rainbow trout. In earlier studies, marine fishes were also found to efficiently retain radiocesium in the gut (Pentreath 1977). For example, 85% of the orally administered ^{137}Cs was removed from the digestive tract by croakers within 6 h (Pentreath 1977). In contrast to ^{137}Cs, AEs of metals in marine fish are generally low, e.g., 10 to 26% for Cd, 4 to 19% for Cr, and 11 to 31% for Zn in the mudskipper *Periophthalmus cantonensis* (Ni et al. 2000). Recently, we also quantified the AEs of ^{137}Cs in marine mussels and a predatory gastropod. The AEs were found to increase with increasing trophic level, e.g., 0.4 to 10% in the mussel *Mytilus edulis*, feeding on diverse food particles, to 44 to 58% in the gastropod *Babylonia formosae habei*, feeding on bivalve tissues (Wang et al. 2000).

Efflux rate constant of radiocesium in the mangrove snapper was rather constant (0.02 d^{-1}) following uptake from the aqueous and dietary phases. The calculated biological retention half-life of radiocesium was about 32 to 36 d. These values were significantly smaller than the biological retention half-lives measured in the freshwater fishes (e.g., years) (Rowan & Rasmussen 1995, Baudin et al. 2000). There are only a few measurements on marine fishes to compare with our data. Depending on different fish species, the calculated biological retention half-lives of radiocesium in marine fishes ranges between 37 and 203 d (Pentreath 1977). It is unknown whether the efflux rate was related to K^+ metabolism or to the high temperature used in our experiments (which may lead to a higher metabolism). For example, the physiological regulation of K^+ in marine fishes may be responsible for the high efflux rate in marine fishes. Cocchio et al. (1995) found that the elimination of radiocesium in the rainbow trout increased by 2× when the ambient K^+ concentration increased from 0.3 to 3.4 mM. The teleost fishes are hypotonic relative to seawater and consequently need to drink seawater to counteract the loss of water from their tissues. The build-up of salt requires the active elimination of salts through gill. Active elimination (vs passive uptake) may be required to remove the salts against the concentration gradient from the tissue to the seawater.

We cannot rule out the possibility of the influence of the higher temperature used in our experiments, typical in the subtropical and tropical waters. Temperature has been demonstrated to be important in affecting the ^{137}Cs efflux in fishes (Cocchio et al. 1995, Rowan & Rasmussen 1995, Peters & Newman 1999). Few studies compared the efflux rate or uptake rate of ^{137}Cs in fishes from tropical waters with temperate or polar species, but some evidence has indicated that the metabolism is different among these regions (Twining

et al. 1996). Jonsson et al. (1999) recently showed that the ecological half-life of radiocesium in freshwater fishes (brown trout and Arctic charr) can be as long as 8 to 22 yr. However, it should be pointed out that the field measurement of radiocesium elimination rate can be confounded by the bi-directional flux of radionuclides (both uptake and elimination) in nature. There was also some considerable difference among different species of fishes in eliminating the radiocesium (Forseth et al. 1998). Furthermore, Garnier-Laplace et al. (2000) indicated that the efflux rate of radio-cesium in trout *Oncorhynchus mykiss* was much higher (0.0170 d^{-1}) following uptake from the water than that measured following uptake from the prey (0.00074 d^{-1}). The mechanism underlying such a difference was not clear from that study.

There is no experimental study on the route of ^{137}Cs exposure in the fish, although it has been speculated that dietary uptake is the dominant process accounting for radiocesium bioaccumulation (Rowan & Rasmussen 1993). Jefferies & Hewett (1971) assumed that >50% of radiocesium in plaice *Pleuronectes platessa* and >80% in ray *Raja clavate* was derived from food uptake. Our model calculation indicated that both water and trophic transfer can be important for the bioaccumulation of ^{137}Cs in predatory fish. The relative importance of the 2 exposure pathways is largely dependent on the concentration factor in the prey organisms as well as the ingestion rate of the fish. Feeding rate has been previously implied to be important in determining the bioaccumulation of ^{137}Cs, including the size dependence of ^{137}Cs radioactivity in fishes (Kasamatsu & Ishikawa 1997). We used a range of feeding rates in our modeling calculation. Feeding rate may be greatly dependent on the seasons and life cycle, and may substantially affect the AE. Garnier-Laplace et al. (2000) therefore introduced the relationship between the ingestion rate and growth rate into the bioaccumulation model. Assuming an average ingestion rate and an average concentration factor in the prey organisms, our calculation would indicate that about 80% of ^{137}Cs in fish would indeed be derived from the dietary source.

The relative importance of different exposure pathways is controlled by the relative magnitude of the influx rates. The uptake rate constant of ^{137}Cs from the aqueous phase was rather constant under different ambient Cs concentrations and within the salinity range of 20 to 30 psu. Thus, the relative importance of dietary exposure is largely a reflection of the variation of fish feeding rate and concentration factors of ^{137}Cs in the prey. It is thus necessary for further studies to focus on the variation of the concentration of ^{137}Cs in prey organisms and the variability of fish feeding rate under different ecological conditions. In contrast to these 2 parameters, the AE, which is critical in the overall influx of radiocesium, did not vary considerably for different fish prey.

Our study has demonstrated that the high AE from an ingested food source is a critical factor contributing to the potential biomagnification of radiocesium in the top marine fish. The TTF is a function of the AE, the ingestion rate of the fish and the elimination rate constant (or efflux) from the animals. The ratio of radiocesium radioactivity in fish to its radioactivity in the lower trophic level is expected to further increase if uptake from the aqueous phase is also considered. The simple kinetic model predicts that, in most circumstances, the TTF is >1 (between 0.9 and 4.4); this strongly implies that ^{137}Cs may be biomagnified in the top marine fishes. At a medium level of ingestion rate (5% of body weight daily), our simple model would predict a TTF factor of 2.0. Consistently, Rowan & Rasmussen (1993) summarized that the bioaccumulation of radiocesium in the piscivorous fishes was generally 2 to 3× relative to the bioaccumulation levels in forage species (planktivores and benthivore fish), despite the fact that the bioaccumulation factor can vary considerably in different systems. They also found that the bioaccumulation factor with a short food chain (e.g., marine system) tends to be lower than those with longer food chains (e.g., freshwater system). In the marine systems, Kasamatsu & Ishikawa (1997) found that the biomagnification factor (^{137}Cs concentration in predator to ^{137}Cs concentration in prey) was about 2.0 (1.8 to 2.2) in the top marine fish. Recently, Garnier-Laplace et al. (2000) and Baudin et al. (2000) estimated a TTF of 1.45 in the top fish predator in freshwater system.

The high trophic transfer is probably most directly related to the high ^{137}Cs AE, despite the lower efflux rate constants than those recorded in the freshwater fishes and the low ingestion rate of fish. In contrast, the AEs of other metals that do not biomagnify in the top trophic level are rather low (Reinfelder & Fisher 1994, Ni et al. 2000). These experimental results demonstrated that radiocesium behaves fundamentally differently from other trace elements, such as Cd and Zn, in marine fish.

Bioaccumulation of radiocesium has been considered to be of less importance in marine systems than in freshwater systems due to the competition with ambient K^+ ions (Rowan & Rasmussen 1993). Thus marine fish are probably less vulnerable to the release of radiocesium than their freshwater counterparts. Nevertheless, the increase in the TTF in the marine predatory fish may be of considerable concern because they are frequently the food items consumed by humans. Furthermore, some marine waters can be noticeably impacted by the fuel reprocessing plants (such as the Sellafield plant in the UK). In these areas it is likely that radiocesium concentration in fishes may be high due to biomagnification.

Our study has therefore demonstrated that the simple kinetic model can be useful in predicting the potential biomagnification in marine food chains. Biomagnification of trace elements has to date been shown only for Hg and Cs. Recently, our study on a marine benthic food chain (phytoplankton to bivalves to gastropod) also indicated that the trophic transfer factor of radiocesium increases with increasing trophic levels and is close to 1 in the top predator (gastropods) (Wang et al. 2000). Biomagnification of ^{137}Cs in top benthic food chain may potentially be likely when the sediment constitutes an important food source for the bivalves due to the high Cs concentration in these sediments compared with its concentration in suspended seston and phytoplankton. In mangrove snappers, the high assimilation efficiency of radiocesium from ingested prey contributes principally to the biomagnification and the significance of dietary exposure to the overall ^{137}Cs bioaccumulation.

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