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The Derivation of Transfer Parameters in the Assessment of Radiological Impacts on Arctic Marine Biota

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ABSTRACT. The initial stage of an environmental impact assessment requires quantification of radionuclide transfer in the study area. This paper evaluates the robustness of the concentration factor (CF) approach in assessing radiological impact on reference Arctic marine biota. By comparing region-specific data sets with recommended generic values for CFs, we tested the hypothesis that transfers to Arctic biota differ from transfers observed in temperate areas for ^{90}Sr, ^{137}Cs, ^{239,240}Pu and ^{99}Tc. Despite the general paucity of data and great uncertainty regarding radionuclide CFs in reference biota, we conclude that the use of Arctic-specific CFs for Sr and Pu can be justified in some cases where differences from generic CFs seem apparent. Where CF data are absent, a biokinetic modelling approach with allometric considerations might be used to bridge data gaps. Such an approach has been used here to estimate the trophic transfer of ^{137}Cs and ^{239}Pu in a marine food chain consisting of four trophic levels. For the simulation concerning ^{137}Cs, the preliminary results suggest that it takes more than five years to attain equilibrium for higher trophic levels (polar cod and harp seal). Biomagnification appears to occur at the lower trophic levels, but not at the highest (seal). For ^{239}Pu, transfer to successively higher trophic levels is low: there is a fall of several orders of magnitude between primary producers, represented by phytoplankton, and polar cod, representing trophic levels 3 and 4. However, the model predicts that this decreasing trend in activity concentrations along the food chain is reversed for the highest trophic level, represented by seal. The simulated results for seal display equilibrium activity concentrations about two orders of magnitude higher than those observed for polar cod (one of its prey species). However, equilibrium (165 years) is not reached during the life span of a seal. The equilibrium ^{137}Cs CFs are approximately 50 l kg^{-1} for zooplankton, 130 l kg^{-1} for polar cod, and 70 l kg^{-1} seal. The predicted equilibrium ^{239}Pu CFs are 2.5 •10^{3} l kg^{-1} for zooplankton and 25 l kg^{-1} for polar cod. For seal, following a one-year equilibration period, a CF of approximately 75 l kg^{-1} is predicted.

Key words: Arctic, concentration factors, biota, dynamic models, radionuclides, uptake, allometric

RÉSUMÉ. Le stade initial d'une étude d'impact environnemental nécessite une évaluation quantitative du transfert de radionucléides dans la zone d'étude. Cet article évalue la robustesse de la méthode du facteur de concentration (FC) pour déterminer l'impact radiologique sur un biote marin arctique de référence. En comparant des ensembles de données spécifiques à une région avec des valeurs génériques recommandées pour les facteurs de concentration, on a testé l'hypothèse selon laquelle les transferts au biote arctique diffèrent des transferts observés dans des régions tempérées pour ^{90}Sr, ^{137}Cs, ^{239,240}Pu et ^{99}Tc. Malgré la pénurie générale de données et un haut niveau d'incertitude concernant les FC des radionucléides dans le biote de référence, on conclut que l'utilisation de FC spécifiques à l'Arctique pour Sr et Pu peut être justifiée dans certains cas où les différences d'avec les FC génériques semblent apparentes. Là où il n'existe pas de données sur les FC, on peut recourir à la modélisation biocinétique tenant compte des éléments allométriques afin de combler les lacunes dans les données. C'est cette approche que l'on a utilisée ici pour estimer le transfert trophique de ^{137}Cs et de ^{239}Pu dans une chaîne alimentaire marine comprenant quatre niveaux trophiques. Pour la simulation relative à ^{137}Cs, les résultats préliminaires suggèrent qu'il faut plus de cinq ans pour atteindre l'équilibre aux niveaux trophiques supérieurs (morue polaire et phoque annelé). La bioamplification semble se produire aux niveaux trophiques inférieurs, mais pas au plus élevé (phoque). Pour ^{239}Pu, le transfert aux niveaux trophiques supérieurs est faible: on constate une baisse de plusieurs ordres de grandeur entre les producteurs primaires, représentés par le phytoplancton, et la morue polaire, qui représente les niveaux trophiques 3 et 4. Le modèle prédit toutefois que cette tendance à la baisse dans l'activité volumique le long de la chaîne alimentaire s'inverse au niveau trophique le plus élevé, représenté par le phoque. Les résultats simulés pour le phoque affichent des activités volumiques à l'équilibre environ deux ordres de grandeur plus élevées que celles observées chez la morue polaire (l'une des espèces-proies du phoque). L'équilibre (165 ans) n'est cependant pas atteint durant la durée de vie du phoque. Les FC de ^{137}Cs à l'équilibre sont environ de 50 l/kg pour le zooplancton, de 130 l/kg pour la morue polaire et de 70 l/kg pour le phoque. Les FC de ^{239}Pu projetés à l'équilibre sont de 2,5•10^{3} l/kg pour le zooplancton et de 25 l/kg pour la morue polaire. Pour le phoque, après une période d'équilibre d'une année, on prédit un FC d'environ 75 l/kg.

Mots clés: Arctique, facteurs de concentration, biote, modèles dynamiques, radionucléides, biomobilisation

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INTRODUCTION

Until recently, the fundamental tenet for the protection of the environment from radiation was based on the premise that if man was protected from harm, then all other components of the ecosystem would be protected (ICRP, 1977, 1991). However, this premise has been increasingly questioned for a number of reasons: it is not always valid; it tends to be inconsistent with environmental protection standards for other hazardous materials; and it is not compatible with a requirement to demonstrate protection of the environment explicitly in line with numerous internationally sanctioned principles and conventions (Pentreath, 1998; Strand et al., 2000). As a result, there have been a number of initiatives, both national (e.g., Amiro, 1997; Copplestone et al., 2001; Jones et al., 2003) and international (e.g., Strand and Larsson, 2001; IAEA, 2002; IUR, 2002; ICRP, 2003), to devise and develop ways of estimating environmental exposures and assessing the subsequent effects of ionizing radiation on flora and fauna. While such initiatives are of international interest, their application to pristine Arctic wilderness areas may be particularly apt. The existence of many ecologically important or vulnerable species and habitats, often key targets for conservation efforts (IAEA, 2002), makes the Arctic region a suitable case for studying the application of an impact assessment system. The European Arctic provides an example of such an environment.

Increasing concern over both potential and actual radioactive contamination in the European Arctic (AMAP, 1998; Bøhmer et al., 2001) is due to the diverse range of nuclear sources in or affecting the region. These sources include power plants, nuclear-powered vessels of the Russian military and civilian fleets, discharges from nuclear reprocessing plants, and sites of weapons tests and nuclear explosions for non-military purposes (Strand et al., 1997). Given the lower biodiversity and the extreme environmental conditions within the region (e.g., low temperature, seasonality in light intensity, ice cover), Arctic ecosystems are potentially more vulnerable to contaminants than organisms in other European climatic regions (AMAP, 1998).

Demonstrating protection of the environment remains problematic. One suggestion has been to use reference organisms to represent the flora and fauna for which radiation doses and potential effects are to be predicted (Pentreath and Woodhead, 2001). The selection of such reference organisms for the European Arctic has been the theme of recent discussions (Beresford et al., 2001; Brown, 2003). Several selection criteria have been applied, including radioecological sensitivity (where in the environment exposure organisms are likely to receive highest internal and external doses), intrinsic radiosensitivity, amenability to research, and the requirement to represent various ecological niches. The final choice of reference organisms, based on these earlier discussions, is presented in Table 1. Furthermore, it became evident that a list or species representing each of these generic organism groups would be useful for several reasons, including a requirement for geometry construction in the process of deriving dose conversion factors. In the context of this study, quantitative information on size, shape, and density are required, and this information can be derived, simply and transparently, from a consideration of real flora and fauna. These representative organisms are also presented in Table 1.

In the initial stage of an environmental impact assessment, the transfer of radionuclides in the environment must be considered. Assessment of radionuclide uptake by marine biota is often based upon the use of equilibrium concentration factors (CF values), which can be defined as the ratio of the activity concentration in the organism (Bq kg^{-1}, normally fresh weight, or f.w.) to concentration in the ambient, which is normally represented by filtered seawater (Bq kg^{-1}, or more practically, Bq l^{-1}). The CF approach has the advantage of being relatively simple and providing the assessor with a large, easily accessible database. Numerous reviews and summaries of various CF values have been made in the past (e.g., IAEA, 1985; Harrison, 1986; Gomez et al., 1991), but the most commonly used, with respect to radiation dose assessments for humans, are the suite of CF values recommended by the International Atomic Energy Agency (IAEA, 1985, in press). Example values from IAEA (in press) of commonly occurring radioactive contaminants for a suite of generic organism groups pertinent to the present study are given in Table 2.

Although the generic organism groups considered by the IAEA (IAEA, 1985, in press) are similar and in some cases identical to the selected Arctic reference organisms, the applicability of these data to the concept of radiological impacts on biota is somewhat limited, as the emphasis in the IAEA review was on estimating exposure to radiation of humans. Hence, data collation in the IAEA reviews focused only on those marine species that constitute food species for humans, normally using information gathered about their edible body parts. When investigating radiation doses to biota, it is important to consider not only those parts of an organism that are normally eaten by humans, but also those body parts that might be of interest from a dosimetric or dose-effects perspective for the organism per se (e.g., the hepatic system, where actinides and other heavy metal radionuclides can accumulate, or the gonads, which are important for fertility). Furthermore, the IAEA-recommended CF values (IAEA, 1985, in press) are generically applicable, whereas radiological protection of the Arctic is conducted in a region where the in situ physical conditions may hypothetically alter transfer to biota (Kryshev and Sazykina, 1986, 1990; Sazykina, 1995, 1998). Cold temperatures are known to alter the metabolic rates of poikilothermic organisms, e.g., fish (see Winberg, 1956). Metabolic rates, in turn, affect the dynamic of contaminant uptake. For example, Kryshev and Ryabov (2000) noted that rates of loss of radiocaesium in fish were comparatively slow in Arctic lakes because of low ambient temperatures.

TABLE 1. Reference organisms for the European Arctic (adapted from Brown, 2003).

Category of organism	Nutritional category	Habitat	Proposed representative organism
Macroalgae	Primary producer	Seashore	*Fucus*
Pelagic crustacean (small crustacean)	Planktotrophic	Pelagic	Northern pink shrimp (*Pandalus borealis*)
Benthic crustacean	Largely detrivorous	Benthic	Kamchatka crab (*Paralithodes camtschatica*)
Mollusc	Largely detrivorous	Benthic	Blue mussel (*Mytilus edulis*)
Pelagic fish (small, planktotrophic)	Planktotrophic	Pelagic	Arctic cod (*Boreogadus saida*)
Pelagic fish (large, piscivorous)	Carnivorous	Pelagic	Atlantic cod (*Gadus morhua*)
Benthic fish	Carnivorous	Benthic	Plaice (*Pleuronectes platessa*)
Mammal	Carnivorous	Islands, coastal areas, ice	Harp seal (*Phoca groenlandica*)
Bird	Carnivorous	Islands	Herring gull (*Larus argentatus*)

TABLE 2. Recommended concentration factors for generic marine organisms (IAEA, in press).

Element	Phytoplankton	Macroalgae	Zooplankton	Mollusca[1]	Crustaceans	Fish
Cs	2×10^1	5×10^1	4×10^1	6×10^1	5×10^1	1×10^2
Tc	4×10^0	3×10^4	1×10^2	5×10^2	1×10^3	8×10^1
Sr	1×10^0	1×10^1	2×10^0	1×10^1	5×10^0	3×10^0
Pu	2×10^5	4×10^3	4×10^3	3×10^3	2×10^2	1×10^2
Am	2×10^5	8×10^3	4×10^3	1×10^3	4×10^2	1×10^2
Po	7×10^4	1×10^3	3×10^4	2×10^4	2×10^4	2×10^3

[1] excluding cephalopods

This work uses the European Arctic as a case study of environmental impact assessment in its initial stage, when exposures must be quantified. The robustness of the CF approach to assessing radiological impact on Arctic marine biota was evaluated in the context of radionuclide uptake by a series of reference organisms. To test the hypothesis stated above—that transfer to Arctic biota differs from that observed in temperate areas—we compared region-specific data sets collated in this study to the recommended values provided by the IAEA (in press). This approach is far from ideal, since the IAEA CF values were designed to apply to all marine environments, and only part of the information used to derive them came from northern marine areas. Nonetheless, it was accepted as being insightful provided that due consideration was afforded the derivation of the IAEA-recommended values. Further considerations are required to determine whether transfer information for generic groups of organisms can appropriately be applied to particular species within the group. Radionuclide transfer data for individual species are normally limited; thus, it is desirable to have the possibility of using surrogate data from related species or a generic group. Finally, the results of the appraisal are critically discussed, with special reference to the use of CF factors in situations where equilibrium may not be assured. Alternative methods for deriving biological transfer data are explored for cases where equilibrium is absent or where empirical data gaps are present.

DATA SETS

An earlier review concerning transfer of radionuclides to generic marine organisms was undertaken by Fisher et al. (1999). The present study has built upon this earlier work, augmenting the database considerably and moreover considering data in the light of their utility within an environmental impact assessment.

In the present study, site-specific radionuclide CF values for Arctic marine samples taken within the Norwegian, Barents, White, Kara, and Greenland Seas biota were collated from extensive literature reviews. CF values (^{90}Sr, ^{137}Cs, ^{239}Pu, ^{240}Pu, and ^{99}Tc) were calculated for Arctic fish, birds, sea mammals, zoobenthos, and macroalgae. For some radionuclide-organism combinations, data for neighbouring sea regions (i.e., the North Sea and North Atlantic) were used because of the scarcity of Arctic-specific data. These data were, of course, not used in any comparisons of Arctic and temperate regions. The collated data set represents the period 1961–99 and was broadly categorized on the basis of selected reference organisms (Beresford et al., 2001; Brown, 2003). In total (across all organisms), 1081 data points were considered for ^{137}Cs, 78 for ^{90}Sr, 118 for ^{239,240}Pu, and 32 for ^{99}Tc. Further details are provided in Kryshev et al. (2002) and Beresford et al. (2003).

From the data collated in this study, estimated CFs were established for the generic reference organisms, and these are presented in Table 3. For the purpose of this analysis, the organism group zooplankton was not considered. Small pelagic crustaceans, though not technically zooplankton, may be considered as an appropriate surrogate for this group. Transfer data have also been collated for species representing each of these organism groups. This information is presented in Table 4.

TABLE 3. Estimated concentration factors for marine organism groups from the European Arctic (l kg^{-1} fresh weight).

Reference organism group	Cs (range)	n	Sr (range)	n	Pu (range)	n	Tc (range)	n
Macroalgae	40 (8–170)	116	100 (40–150)	14	300 (800–34 000)	46	40 000 (1000–50 000)	18
Mollusc	40 (10–80)	31	n.a.[1]	0	150 (20–260)	10	300 (100–600)	5
Crustacean	50 (10–150)	41	15 (10–20)	7	300 (40–500)	8	4000 (300–9 300)	8
Fish	80 (40–1 800)	630	10 (3–15)	37	130 (120–900)	23	?	1
Sea bird	300 (50–7 000)	55	n.a.	0	<100–200	6	n.a.	0
Sea mammal	200 (50–600)	175	1 (0.7–1.5)	17	< 400 (20–700)	15	n.a.	0

[1] n.a. = no data available.

TABLE 4. Estimated concentration factors for "representative" marine species from the European Arctic (l kg^{-1} fresh weight).

Reference species	^{137}Cs	^{90}Sr	^{99}Tc	^{210}Po	^{239,240}Pu	^{241}Am
Cod	80 ± 40	15 ± 10	–	600	140 ± 60	–
Plaice	100 ± 30	8 ± 5	–	5330	< 200	–
Polar cod	100 ± 50	5 ± 3	–	3330	< 200	–
Mussel (soft tissues)	50 ± 14	–	300 ± 200	6.0×10^4	150 ± 110	2.0×10^4
Crab (muscles)	150 ± 40	15 ± 5 (Crustaceans)	1400 ± 400	$(3.7 \pm 1.5) \times 10^4$	300 ± 200 (Crustaceans)	500 (lobster)
Shrimp (muscles)	35 ± 11	15 ± 5 (Crustaceans)	100	$(4.5 \pm 0.5) \times 10^4$	300 ± 200 (Crustaceans)	–
Gull	580 ± 200	–	–	–	100 ± 50	–
Harp seal	70 ± 20	10 ± 5	–	$(2.1 \pm 0.3) \times 10^4$ (sea mammals, muscles)	400 ± 300 (sea mammals)	–

CONCENTRATION FACTORS

Overall, several tentative conclusions can be drawn about differences between radionuclide uptake in Arctic environments and global radionuclide uptake averages. A more rigorous statistical analysis to test for differences between data sets (e.g., a Student's t test or Mann-Whitney U test) has not been possible owing to a lack of information on the derivation of IAEA-recommended values. The analyses are, therefore, of a qualitative nature only.

^{137}Cs CF values for macroalgae, molluscs, crustaceans, and fish from the Arctic seas are not significantly different from corresponding generalized world values. This result slightly contradicts the observation made by Fisher et al. (1999), based on a smaller data set, that CFs, including those for radiocaesium, are somewhat higher in organisms in the Arctic than in organisms in temperate waters (based on IAEA, 1985). They emphasized, however, that variability in data was large.

The fact that IAEA (in press) has recently revised upward its recommended ^{137}Cs CF values for molluscs and crustaceans may partly explain this discrepancy between reviews, although the ^{137}Cs CF values derived in the present study are generally lower than those reported in Fisher et al. (1999). IAEA (in press) now recommends CF values for ^{137}Cs of 400 for pinniped muscle and 300 for cetacean muscle. As these data were partly derived using northern or Arctic sea data, no definitive conclusion can be drawn from their observed similarity with the CF values derived in the present study.

^{90}Sr CF values in macroalgae, crustaceans, and fish from the Arctic seas appear to be somewhat higher than global average values (cf. Tables 2 and 3). Similar observations were made by Fisher et al. (1999), as noted above. In the case of macroalgae, however, caution is required in interpreting this observation. The IAEA-recommended value is biased towards edible seaweeds, of which green and red seaweeds constitute important components. The facts that CF data for macroalgae in Arctic regions are based solely on brown seaweeds and that brown algae are unique in accumulating strontium (Bowen, 1979) may alone account for this apparent difference in uptake.

^{239,240}Pu CF values for crustaceans and fish are similar to those values derived from world ocean data (IAEA, in press), but macroalgae and molluscs from Arctic environments both appear to exhibit CFs distinctly lower than those from temperate environments. In the case of macroalgae, however, the range observed for Arctic data was extremely large (800–34 000), with numerous CF values above the world-ocean recommended value. It is not possible, therefore, to establish whether CFs are significantly different from this basic appraisal. In the case of molluscs, difference between the data sets is more apparent because even the maximum value in the Arctic data set falls below the world-ocean recommended value. A speculative reason can be provided to account for the discrepancy. The IAEA-derived values are from the English Channel, which receives discharge from the nuclear reprocessing facility at La Hague (Normandy, France). Given the large suspended loads associated with output from this high-energy environment, we might expect a significant proportion of Pu present in the water column to be associated with particulate material—and therefore available to particulate-feeding benthos, such as molluscs. In contrast, Arctic studies have shown that most of the plutonium in Arctic shelf seas is associated with fully dissolved or low molecular weight fractions (Mitchell et al., 2002), forms that might exhibit quite different bioavailability (defined in terms of potential uptake by filter-feeding benthos).

Comparison of ^{99}Tc CF values between Arctic and temperate regions is possible only for molluscs and macroalgae. The limited data available suggest that differences in CFs between regions are small, possibly insignificant. For macroalgae, the range expressed by CF values is very large, which may reflect interspecies variations in ^{99}Tc uptake (McCartney and Rajendran, 1997). For all other organism groups, Arctic region data sets were not extensive enough to allow even a cursory comparison to temperate regions. For seabirds, few, if any, CF data have been published for temperate regions rendering any comparison exercise impossible.

CF data for organism groups can be compared with those for representative organisms (cf. Tables 2 and 3 vs. Table 4). The available data provide no strong evidence to suggest that CF values for individual species are strikingly different from CF values for the group as a whole. This is not a surprising result in view of the fact that individual species data are used to derive estimated, or recommended, organism group CF values. Nonetheless, we advise caution in the application of generic values in any impact assessment context. A case in point is provided by the uptake of ^{99}Tc to the generic group crustaceans. Within this group, lobsters (e.g. *Homarus gammarus*) exhibit much higher CFs than, for example, edible crabs (*Cancer pagarus*) because of the very high affinity of the lobster's green gland ("tomalley") and hepatopancreas for this radionuclide (Masson et al., 1989; Busby et al., 1997; Olsen and Vives i Batlle, 2003). This difference in uptake of Tc may be related to physiological specialization whereby phylogenetically more primitive forms exhibit higher technetium bioaccumulation than more advanced forms, although such speculation generates no information on the underlying physical mechanisms responsible for the wide differences in empirically derived CFs (Swift, 1989). One might conjecture that the application of a single generic organism group CF value might lead to an uncertainty of up to several orders of magnitude in exposure estimates for individual species.

In the context of environmental impact assessments, there is a lack of data on radionuclide distributions within actual organisms, i.e., CF data for specific organs. Such information might be critical for accurate determination of the actual dose or dose-rate that an animal is receiving, especially in cases where a radionuclide is accumulated by certain organs. A case in point is, again, provided by ^{99}Tc, which is known to be accumulated by the green gland of lobster at a level approximately two orders of magnitude higher than that observed in muscle (Busby et al., 1997). It is difficult to estimate the biological effects of exposure on individual organs because most data on dose effects pertain to whole-body exposure from external radiation sources (UNSCEAR, 1996).

The set of estimated CF values for ^{137}Cs for fish (cod, *Gadus morhua*), sea mammals (harbour seal, *Phoca vitulina*), and macroalgae (*Fucus vesiculosus*) displays an obvious time dependence (Fig. 1), reflecting the relatively slow response of organisms to ambient seawater concentrations. The fact that marine biota ^{137}Cs uptake processes did not reach equilibrium over the long observational time periods considered in this study clearly illustrates the limitations associated with the use of the equilibrium CF approach in marine models and radiological impact assessments.

The CF approach is open to criticism because:

1) it provides no information concerning the types of processes or mechanisms in operation during biological uptake, or information regarding the chemical or physical speciation of the radionuclide;
2) the relationship between the radionuclide concentration in water and within (the organs or whole body of) a high–trophic level organism that derives most of its contaminant load from ingested food may not be a simple, linear one;
3) the assumption that the system is under equilibrium, a requirement for CFs to be truly applicable, is often invalid;
4) Even if the generic data for the world oceans (from IAEA, in press) are employed, and the limitations on use considered above are accepted, the uptake of many radionuclides to certain reference organism types is poorly, if at all, described. A good example can be presented for sea mammals and birds, for which data coverage extends to only a handful of radionuclides and where the great preponderance of data exists for ^{137}Cs.

In a comprehensive review on this theme, Coughtrey and Thorne (1983:496) concluded that "the use of one concentration factor for either marine organisms in general, for the same organism in different sites, for studies involving chronic compared to acute contamination, for short-lived compare to long-lived nuclides, …for open-ocean compared to coastal sites, and for specific animal tissues compared to whole animals, is highly unsatisfactory."

Dynamic Modelling

Other approaches to modelling the transfer of radionuclides in ecological systems will therefore be explored in this section. Biokinetic models may allow us to make more realistic prognoses concerning the dynamic response of an ecological system and to derive tentative estimates concerning equilibrium CFs. Where data are lacking on some of the parameters required for simulation, allometric relationships may provide surrogate values. The allometric approach is based on the observation that many metabolic parameters, including basal metabolic rates, ingestion rates, and biological half-lives, are proportional to the size of an organism. Such approaches have been applied elsewhere for the very purpose of deriving transfer data where empirical data sets are unsatisfactory (U.S. Department of Energy, 2002; Higley et al., 2003). To

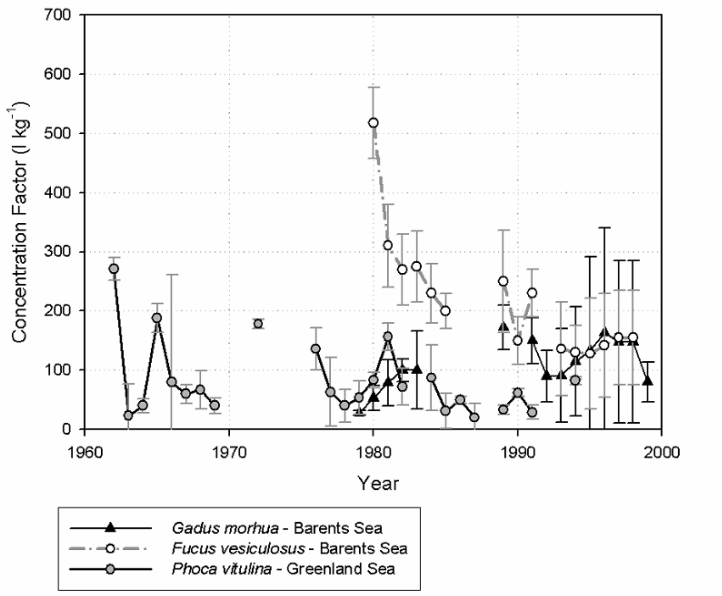

FIG. 1. Temporal variation in ^{137}Cs CF values for Atlantic cod (*Gadus morhua*) in the Barents Sea (annual average 1 kg^{-1} f.w. ± SD), bladderwrack (*Fucus vesiculosus*) in the Barents Sea (annual average d.w. ± SD), and common seal (*Phoca vitulina*) in the Greenland Sea (annual average 1 kg^{-1} f.w. ± SD).

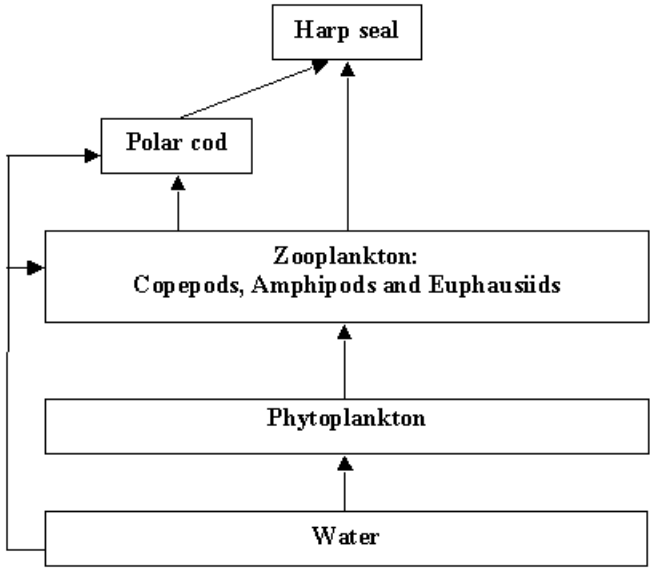

FIG. 2. Food-chain model for harp seal in the Barents Sea, simplified from Dommasnes et al. (2001).

demonstrate how this type of model might be employed to fill the required knowledge gaps, we developed a simple food-chain model based on the work of Dommasnes et al. (2001) and information in the open literature, to consider the transfer of selected radionuclides (^{137}Cs and ^{239,240}Pu) to reference organisms in the pelagic food chain phytoplankton-zooplankton-polar cod-harp seal (Fig. 2).

The model, based on the work of Thomann (1981), Landrum et al. (1992), and Fisher (2002), considers uptake via food and water for aquatic organisms. Excretion/elimination rates are assumed to be independent of the uptake route, the assimilation efficiency is assumed to be independent of food type, and predators are assumed not to assimilate the activity concentration in gut content of their prey. Further assumptions are that the phytoplankton and zooplankton (trophic levels 1 and 2) are homogeneous groups, described by specified parameter values rather than by ranges, and that the growth rate for all organisms is 0. This last assumption may be a particularly poor one (Thomann, 1981), but the complexity of the weight dynamics for the organisms in question would require further, more detailed study.

The time-dependent transfer of radionuclides within the food chain can be described by simple, first-order differential equations, one for each trophic level.

Trophic Level 1: Phytoplankton (equilibrium with water concentration):

$$C_p = CF \cdot C_w \qquad (1)$$

where C_p is the radionuclide activity concentration in phytoplankton (Bq kg^{-1} f.w.); CF is the bioconcentration factor for phytoplankton (l kg^{-1}); and C_w is the radionuclide activity concentration in sea water (Bq l^{-1}).

Trophic Level 2: Zooplankton (uptake via water and food):

$$\frac{dC_z}{dt} = AE_z \cdot IR_z \cdot C_p + k_{uz} \cdot C_w - C_z \cdot k_{ez} \qquad (2)$$

where AE_z is the assimilation efficiency (dimensionless) for zooplankton; IR_z is the ingestion rate per unit mass of zooplankton (kg f.w. d^{-1} per kg f.w.); C_p is the activity concentration in phytoplankton (Bq kg^{-1} f.w.); k_{uz} is the rate of radionuclide uptake by zooplankton directly from the water column (d^{-1}); C_w is the activity concentration in water (Bq l^{-1}); C_z is the activity concentration in zooplankton (Bq kg^{-1} f.w.); and k_{ez} is the excretion rate from zooplankton (d^{-1}).

Trophic Level 3: Polar cod (uptake via water and food):

$$\frac{dC_{pc}}{dt} = AE_{pc} \cdot IR_{pc} \cdot C_z + k_{upc} \cdot C_w - C_{pc} \cdot k_{epc} \qquad (3)$$

where AE_{pc} is the assimilation efficiency (dimensionless) for polar cod; IR_{pc} is the ingestion rate per unit mass of polar cod (kg f.w. d^{-1} per kg f.w.); k_{upc} is the rate of radionuclide uptake by polar cod directly from the water column (d^{-1}); C_{pc} is the activity concentration in polar cod (Bq kg^{-1} f.w.); and k_{epc} is the excretion rate from polar cod (d^{-1}).

Trophic Level 4: Harp seal (uptake via food only):

We assume that the uptake of radionuclides directly from the water column to the harp seal is negligible and that the harp seal's diet, in simplified terms, consists of 50% polar cod and 50% zooplankton.

$$\frac{dC_{hs}}{dt} = 0.5 \cdot \left(AE_{hs} \cdot IR_{hs} \cdot C_z\right) + 0.5 \cdot \left(AE_{hs} \cdot IR_{hs} \cdot C_{pc}\right) - C_{hs} \cdot k_{ehs} \qquad (4)$$

where AE_{hs} is the assimilation efficiency (dimensionless) for harp seal; IR_{hs} is the ingestion rate per unit mass of harp seal (kg f.w. d^{-1} per kg f.w.); C_{hs} is the activity concentration

TABLE 5. Feeding parameters used in the modelling process (after Thomann, 1981).

Organism (Trophic level)	Assumed weight range (g f.w.)	Food consumption (kg d^{-1} per kg f.w.)
Zooplankton (2)	0.001–1	0.105
Small fish (3)	0.005–50	0.017
Large fish (4)	5–5000	0.009

in harp seal (Bq kg^{-1} f.w.); and k_{ehs} is the excretion rate from harp seal (d^{-1}).

From studies conducted under laboratory conditions, it is assumed that the uptake of actinides by phytoplankton cells reaches equilibrium with their ambient media within a few days (Fisher et al., 1983). This is also true for other actinides, including Am, Cf, and Np. This supports (at least partially) our simplifying assumption at the basis of the model, i.e., that equilibrium between seawater and phytoplankton occurs instantaneously.

Parameterization of the biokinetic allometric model is a topic requiring some attention. For lack of more detailed information, the generic values reported in IAEA's Technical Report 247 (IAEA, 1985) have been used for the radionuclides considered. CF values of 20 for Cs and 1×10^5 for Pu have been reported in phytoplankton.

Food consumption or ingestion rates (normalized to the f.w. of the organism) have been tabulated by Thomann (1981) for different trophic levels (Table 5). Polar cod has been defined as a large fish, although in reality the species probably intersects trophic levels 3 and 4 as defined by Thomann (1981). Adult polar cod may attain lengths of up to 40 cm and weigh several hundred grams.

Innes et al. (1987) have provided the following allometric relationship for the ingestion rate, IR (kg f.w. d^{-1} per kg f.w.), for adult seals:

$$\mathrm{IR} = 0.079M^{0.71} \tag{5}$$

where M is the weight of the seal (kg).

Assuming a seal weighing 160 kg (a reasonable estimate of the adult weight of a harp seal (*Phoca groenlandica*) the derived (weight-normalized) ingestion rate is 0.018 kg f.w. d^{-1} per kg f.w. Radionuclide-specific parameters defining uptake rates from water, excretion rates and assimilation efficiencies for zooplankton and fish are presented in Table 6. The parameter values for trophic level 3, large fish, have been taken to be representative of polar cod.

For the seal, assimilation efficiencies for both ^{137}Cs and ^{239}Pu have been set to the same value, that representative of lower levels in the food chain. Direct radionuclide uptake from the water column is assumed to be zero.

An allometric relationship may be used to estimate the ^{137}Cs excretion rate for seal. The following equation has been applied by the U.S. Department of Energy (2002) based on earlier studies (Whicker and Shultz, 1982):

$$\lambda_i = \frac{1\mathrm{n}2}{3.5M^{0.24}} \tag{6}$$

TABLE 6. Input parameters for ^{239}Pu and ^{137}Cs (after Thomann 1981).

Trophic level	Plutonium-239			Caesium-137		
	k_u	k_e	AE	k_u	k_e	AE
Zooplankton (2)	18.7	0.05	0.01	0.49	0.03	0.5
Small fish (3)	0.3	0.02	0.01	0.07	0.003	0.5
Large fish (4)	0.01	0.01	0.01	0.01	0.0018	0.5

where λ_i is the biological decay constant (d^{-1}); and M is the mass of the animal (g f.w.).

Equation (6) yields an excretion rate of 0.0112 d^{-1} for seal. Although this value has been used in this preliminary version of the model, it is apparent that using elimination rates based on allometric relationships leads to a more rapid than expected loss from high-level predators such as seal. For example, applying an allometric relationship for man with assumed mass of 70 kg yields an excretion rate of 0.0136 d^{-1}. This is somewhat greater than expected from the application of a more complex elimination model: i.e., if the elimination rate for man (e.g., ICRP, 1979) is taken, the long component of elimination is at a rate of 6.3×10^{-3} d^{-1}. The data in Table 6 suggest that the excretion rate decreases as trophic level increases, although this trend may be offset because mammals are homoeothermic, with concomitantly higher metabolic rates (for a stated mass). More work is required in deriving more robust excretion rate data for radiocaesium.

Similarly, a biological half-life can be derived for Pu using a simple allometric relationship. This relationship is defined as (U.S. Department of Energy, 2002):

$$\lambda_i = \frac{1\mathrm{n}2}{0.8M^{0.81}} \tag{7}$$

and yields an excretion rate of 5×10^{-5} d^{-1} for a 160 kg seal (the mass is entered in Equation 7 in units of grams).

However, this allometric relationship requires further investigation. The uptake and translocation of Pu are complex and depend on a number of factors, including the age of the mammal. Variable removal rates are likely to be associated with different tissues (e.g., blood, muscle, bone, etc). Retention equations have been developed for humans (see ICRP, 1988) and may be appropriately applied to model depuration for some other mammals.

The model was constructed and equations solved numerically using the modelling software "ECOLEGO" (Avila et al., 2003) in a Matlab© environment, with ^{137}Cs and ^{239}Pu water activity concentrations set to unit concentrations and radioactive decay from each compartment included. Simulation results of the biokinetic allometric model for ^{137}Cs and ^{239}Pu are shown in Figures 3 and 4, respectively.

For ^{137}Cs, the results from model runs suggest that equilibrium is not attained for higher trophic levels—polar cod and harp seal—until 2000 days after initial contamination. This result has obvious implications in relation to the interpretation of field data if activity concentrations in

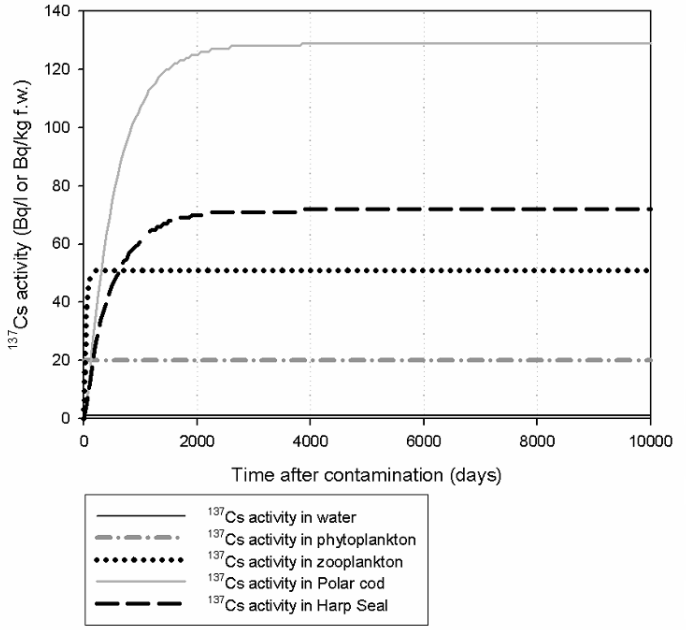

FIG. 3. Predicted whole-body ^{137}Cs activity concentrations (fresh weight) for selected marine organisms derived from the biokinetic allometric model.

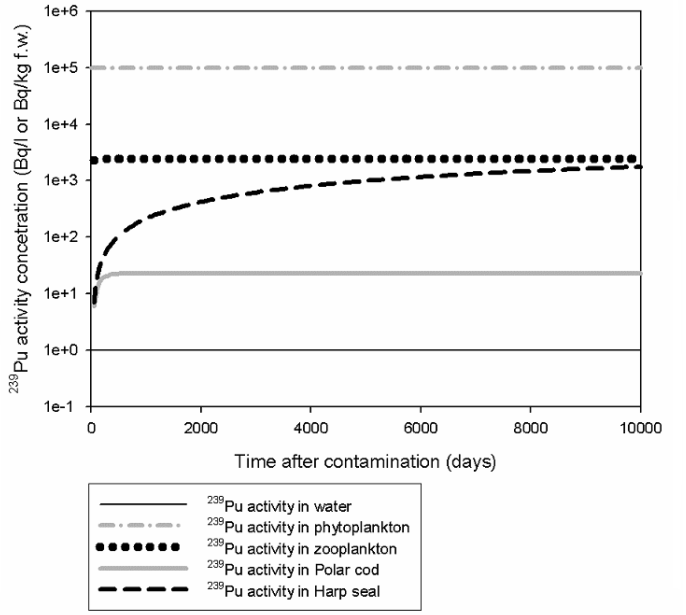

FIG. 4. Activity concentrations (fresh weight) of ^{239}Pu for selected marine organisms derived from biokinetic allometric modelling. Results for polar cod and seal are for the whole body.

water are changing rapidly with time. Biomagnification (increase in body mass concentration of a contaminant as it passes from low trophic levels to higher ones) appears to occur at the lower trophic levels, but not at the highest trophic level, i.e., seal. It should be noted that dietary assumptions also affect the concentrations in seal. If a fish-only diet is assumed, a harp seal CF in excess of approximately 105 can be derived. However, the uncertainty associated with the excretion rate of ^{137}Cs for seal is large, and this parameter has a significant effect on the equilibrium CF. Setting the ^{137}Cs excretion rate to 0.0018 (Table 6) results in a CF of several hundred for seal. Equilibrium ^{137}Cs CF values are approximately 50 for zooplankton, 130 for polar cod, and 70 for seal. These values appear sensible. They compare well with the recommended values (IAEA, in press) of 40 for zooplankton and 100 for generic fish. The ^{137}Cs value of 70 for seal corresponds directly to the empirically derived value included in Table 4 for harp seal.

Several points of interest arise from the simulation for ^{239}Pu (Fig. 4). Transfer of ^{239}Pu to successively higher trophic levels is low, with a decrease of several orders of magnitude observed between phytoplankton (representing primary producers) and polar cod (representing trophic level 3–4). However, the model predicts that this decreasing trend in activity concentrations along the food chain is reversed for the highest trophic level, represented by seal. The simulated results for seal display activity concentrations in the region of two orders of magnitude higher than those observed for polar cod (one of its prey species), once the system has equilibrated. This prediction is strongly influenced by the other component of the seal's diet, zooplankton, which has a high activity concentration associated with it. Equilibrium is attained very slowly for seals (reflecting in part, the very low, allometrically derived excretion rate). In this case, equilibrium is only truly obtained after 6×10^4 days (165 years) of simulation. Clearly, even in the unlikely circumstance that water concentrations remain unchanged over highly protracted time scales, equilibrium is unlikely to be attained over the lifetime (in the order of decades) of the seal.

The equilibrium Pu CF values of 2.5×10^3 (zooplankton) and 25 (polar cod) predicted from model runs, compare favourably with values recommended by IAEA (in press) of 4×10^3 for zooplankton and 100 for generic fish. For seals, as discussed above, a true equilibrium CF value of 4.5×10^3 between the water and body compartments is not obtained over the lifetime of the organism. However, following a five-year equilibration time (although this value is arbitrarily chosen, it represents a "realistic" contaminant-biota contact period), a concentration ratio value of circa 390 was predicted. This latter value compares well with the empirically derived value presented in Table 4 of 400 ± 300. In view of the discussions presented here, the appropriateness of applying a Pu CF value to a high-level predator, such as a seal, is clearly open to question.

CONCLUSIONS

Given the limited data available and the problems associated with compatibility of generic and Arctic data sets, little can be concluded about the effect of Arctic environmental conditions on radionuclide uptake. A general paucity of pertinent data and large uncertainties characterize the CF database. In numerous cases, no information is available at all. In light of these observations, it might be concluded that no recommendation can be made to apply

Arctic-specific CF values instead of generic values, in most instances. However, for some radionuclides, distinct differences are apparent. In the case of Sr, for example, Arctic CFs for fish and crustaceans appear to be higher than corresponding world-generalized values. For Pu uptake to molluscs, Arctic values are distinctly below those recommended for generic application. In such cases, using region-specific CF data might be justified. In the context of environmental impact assessments, it is also noteworthy that data pertaining to uptake to specific organs are very poorly characterized. Such data may be crucial in the derivation of robust exposure (i.e., dose-rate) estimates.

The numerous limitations associated with CFs, not least the fact that environmental compartments are rarely under equilibrium following non-uniform radionuclide inputs, renders the application of dynamic models desirable. Furthermore, such models may help to fill numerous gaps in the radionuclide transfer data for many biota types. In the present study, the application of a multi-compartmental model, parameterized using allometrically derived values where appropriate, has allowed the derivation of Cs and Pu CFs for several marine trophic levels. The preliminary estimates agree well with empirical data sets and demonstrate that, in some cases, the application of an equilibrium CF is highly inappropriate. An example is the simulated outputs for Pu in "seal," which take 165 years to attain equilibrium following a uniform activity concentration in the water compartment. In this case, a concentration ratio selected for a defined time interval (commensurate with realistic contaminant-biota contact times) might be more sensibly applied.

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