

UCRL-JRNL-211213



LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

137Cs and 210Po in Pacific Walrus and Bearded Seal from St. Lawrence Island, Alaska

T. F. Hamilton, D. J. Seagars, T. Jokela, D.
Layton

April 8, 2005

Marine Pollution Bulletin

DISCLAIMER

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

^{137}Cs and ^{210}Po in Pacific Walrus and Bearded Seal from St. Lawrence Island, Alaska

Terry Hamilton^{1,*}, Dana Seagars², Terry Jokela¹, and David Layton¹

¹Environmental Science Division, Lawrence Livermore National Laboratory, P.O. Box 808, Livermore, CA 94550-0808, USA

²U.S. Fish and Wildlife Service, Marine Mammals Management Field Office, 1011 East Tudor Road, Anchorage, AK 99503, USA

Abstract

The activity concentration of Cesium-137 (^{137}Cs) and naturally-occurring Polonium-210 (^{210}Po) were measured in the muscle tissue, kidney and liver of Pacific walrus (*Odobenus rosmarus divergens*) and bearded seal (*Erignathus barbatus*) collected by native hunters from the Bering Sea. The mean ^{137}Cs concentrations in muscle, liver and kidney of Pacific walrus were 0.07, 0.09 and 0.07 Bq kg⁻¹ (N= 5, wet weight), respectively, and 0.17, 0.10, and 0.17 Bq kg⁻¹ (N=2, wet weight), respectively, in bearded seal. In general, ^{137}Cs tissue concentrations are significantly lower than those previously reported for mammals from other regions. By comparison, ^{210}Po activity concentrations appear to be higher than those reported elsewhere but a larger variation. The mean ^{210}Po concentration in the muscle tissue, liver and kidney of Pacific walrus (N=5, wet weight) were 28.7, 189, and 174 Bq kg⁻¹, respectively. This compares with ^{210}Po concentration values (N=2, wet weight) of 27, 207, and 68 Bq kg⁻¹ measured in the muscle tissue, liver and kidney, of bearded seal, respectively. Estimated bioaccumulation factors—as defined by the radionuclide concentration ratio between the target tissue to that in sea water—were two to three orders of magnitude higher for ^{210}Po than those of ^{137}Cs . We conclude from radiological dose estimates that ingestion of ^{137}Cs in foods derived from walrus and seal will pose no threat to human health. This work has important implications for assessing health risks to Alaskan coastal communities concerned about the dumping of nuclear waste in the Russia Arctic.

Keywords: ^{137}Cs , ^{210}Po , Bering Sea, marine mammals

* Corresponding author, e-mail: hamilton18@llnl.gov

Introduction

Cesium-137 (^{137}Cs) is one of the most abundant anthropogenic radionuclides produced during atmospheric nuclear weapons testing programs (1945-81) and deposited into the world's ocean (Hamilton, 2004). ^{137}Cs also forms an important constituent of nuclear waste inventories dumped in the Kara Sea and the North-Western Pacific Ocean (Layton *et al.*, 1997a) and at contaminated sites in the North Siberian Basin (Bradley and Jenquin, 1995). Moreover, radioactive contaminants derived from Russian Arctic regions may be transported along the Siberian continental shelf and slope, and eventually impact on North American waters. The focus of this study was to provide high quality baseline data on radionuclide concentrations in marine mammals as a basis for conducting a risk assessment on Alaskan coastal communities exposed to radionuclides in subsistence diets. Potential dietary exposures to ^{137}Cs have also been compared with those derived from polonium-210 (^{210}Po)—a naturally occurring radionuclide and an important contributor to the dose to man through consumption of marine foods (Pentreath and Alington, 1988). The concentration of any contaminant in a marine organism reflects the sum of all bioaccumulation, metabolic, and elimination processes affecting the transfer of the contaminant to a whole organism and/or target tissue. The main source of ^{137}Cs in the Pacific Ocean can be attributed to worldwide fallout deposition from atmospheric nuclear weapons testing (Hamilton, 2004). A large fraction of the ^{137}Cs oceanic deposit has been retrained in surface waters with latitudinal and/or localized inputs becoming more obscure over time (Hamilton *et al.*, 1996). Consequently, marine mammals living within a defined region such as the Bering Sea have been exposed to near steady-state ^{137}Cs conditions (food + water) from which field-derived radionuclide concentration factors (CF) may be calculated. Similarly, field-derived CF values can be estimated for ^{210}Po and/or other radionuclides, and the results used to determine the criticality

of different radionuclides and/or exposure pathways in contributing to the risk to populations from potential transfers or releases of radionuclides into the marine environment. Over the past decade there has been a large international effort under the Arctic Monitoring and Assessment Programme (AMAP) to assess the state of the Arctic environment with respect to pollution issues (AMAP, 1998a) including those pertaining to dumping of nuclear waste and other sources of marine radioactivity in the Arctic Ocean. Samples of fish, seals and whales collected in Greenland and Icelandic waters have been analyzed for ^{137}Cs since the early 1960s (Rissanen *et al.*, 1997; AMAP, 1998b) but precise measurements of radionuclide concentrations in tissues of marine mammals from the Arctic regions of North America are relatively few. Moreover, recommended CF values used in radiological assessments are often based on extrapolated average values taken over broadly characterized groups (e.g., mammals, fish, mollusks, etc.) and/or geographical regions, and may not be directly applicable to coastal communities of Alaska.

The animals used in this study—five individuals of Pacific Walrus (*Odobenus rosmarus divergens*) and two individuals of Bearded Seal (*Erignathus barbatus*)—were caught by Alaskan hunters for food. Pacific walrus primarily consume bivalve mollusks, although they are known to feed on a wide variety of benthic and epibenthic invertebrates such as gastropods, polychaetes, and molting crabs; also demersal fishes and other pinnipeds may be consumed (Fay *et al.*, 1977; Fay, 1982; Fay and Stoker, 1982a,b; Lowry *et al.*, 1980; Fay *et al.*, 1984.). Bearded seal eat a diversity of epifaunal and infaunal invertebrates such as crabs, shrimp, clams, and snails, as well as demersal and pelagic fish such as capelin (*Mallotus villosus*), Arctic cod (*Boreogadus saida*), and scuplins (Burns, 1967; Lowry *et al.*, 1979; Finely and Evans, 1983; Antonelis *et al.*, 1994). Therefore, dietary habits of bearded seal and Pacific walrus alone

may lead to real differences in the body burden of radionuclides between the two species, and from those of more pelagic and piscivorous feeding marine mammals. Furthermore, food consumption of seal and walrus (and thus, exposure to a particular contaminant) in the Bering and Chukchi Seas varies with their seasonal distribution (Burns and Frost, 1979; Lowry *et al.*, 1980; Fay *et al.*, 1984;). Both species move throughout the relatively shallow (<100 m) waters of the Beringean platform as the pack ice advances and recedes each season (Fay, 1982; Fedeseev, 1990;) but utilize different forms and types of ice (Fay 1974; Braham *et al.*, 1984). With the break-up and retreat of the pack ice in spring, the majority of the Pacific walrus population migrates from the Bering Sea into the Chukchi Sea, returning in the fall to over-winter. About 25-50% of the sub-adult and adult male walrus forego the northward migration, remaining on summer haul-out sites along the Russian Bering Sea coast and in Bristol Bay, Alaska (Gilbert *et al.*, 1992). Bearded seals are commonly found along the southern extent of the pack ice in the Bering Sea during winter, and through spring into summer follow the southern margin of the retreating pack ice in the northern Chukchi Sea. The authors recognize that the value of this study is limited by the small population size. However, the high quality of the data coupled with our knowledge of the feeding habits and seasonal distributions of mammals in the Bering and Chukchi Seas do provide a meaningful basis for evaluating trends in levels of radionuclide contaminations between species, and in providing reliable predictions about potential impacts of radionuclide releases on coastal communities concerned about exposures to radioactive contaminants associated with the dumping of nuclear waste in the Arctic Seas.

Sections of muscle, liver and kidney from each animal were collected under the careful supervision of personnel from the U.S. Fish and Wildlife Service—Marine Mammals Management Field Office and the samples transported to the Lawrence Livermore National Laboratory (LLNL) for

analysis. Here we present the results of ^{137}Cs and ^{210}Po analyses on muscle tissue, liver and kidney. Our data are used to estimate field derived Concentration Factor (CF) values—as defined by the radionuclide concentration ratio between the whole organism or target tissue to that in seawater—and the results compared with those CF values used in dose assessment models. Consequently, our baseline CF values could be used with predicted concentrations of radionuclides in North American waters to provide a better insight into the potential health and ecological impacts of radionuclide releases in the Russian Arctic on Alaskan coastal communities.

Materials and Methods

Sample Collection & Storage

Tissue sections were collected from Pacific walrus and bearded seal during May 1996. Alaskan hunters caught the animals by off St. Lawrence Island in the Bering Sea (Fig. 1) as part of their normal subsistence hunting practices. Meristics, sex, morphological as well as other data relevant to the health and maturity of walrus and seals were collected within minutes of post-mortem. Tissues were sampled within 30 minutes of death by working cooperatively with native hunters as they butchered animals for meat. Up to 10 kg of muscle tissue, and 1-4 kg chunks of kidney, liver, and "coak" (skin with some underlining muscle/fat) were collected for radionuclide measurements, and each sample immediately placed in labeled plastic bags. Sample collection was standardized as follows: muscle—trapesius and latissimus groups, kidney—whole (bearded seal, most walrus) or a nearly whole large chunk, liver—large section taken from the central lobe, and coak—sternal region between anterior and posterior margins of the fore flippers. Samples were held in a cooler at near freezing temperature during transport (ca. 2-4 hours) from sea ice to field quarters. Frozen samples

were subsequently shipped to the Lawrence Livermore National Laboratory (LLNL) for immediate analysis. Tissue samples (~1 cm³) for subsequent histological examination were also collected within 30 minutes post-mortem, and placed in 10% buffered formalin; analysis was conducted by the Armed Forces Institute of Pathology (Department of Veterinary Pathology, Washington, D.C.). For walrus, two canine teeth were collected for subsequent laboratory age determination following the procedures described in Seagars *et al.*, (1995) and Fay *et al.*, (1986). For bearded seals, standard lengths were taken and the lower jaw (for teeth) and claws were collected where possible for subsequent age determination following procedures of Bengston and Siniff (1981), Burns and Frost (1979), Burns (1967), Scheffer (1967), Laws (1962), and McLaren (1958).

At LLNL, the tissue samples were partially thawed and either coarsely ground in a meat mincer or diced into small pieces. The samples were then packed in plastic cartons, the sample cartons placed in a large capacity freeze drying unit, and the material dried to a constant weight over a period of 5 to 6 weeks. Each sample was then reconstituted into a single bulk sample and homogenized in a laboratory blender.

Determination of ¹³⁷Cs

Up to 2 kg of dry, homogenized material was used for each analysis. Each sample was dry ashed at 450°C for 4-5 days, and then leached with concentrated HNO₃ at 70°C for 3-4 hours. Any remaining insoluble material was removed by filtration. Ten mg of stable Cs was then added to the sample leachate (as a chemical yield tracer) and ¹³⁷Cs (Cs) separated from the bulk solution by batch extraction onto 1-2 g of the microcrystalline cation exchanger, ammonium molybdophosphate (AMP), at pH 2-3. The precipitate was then recovered by centrifugation and dissolved in a minimum quantity of 10 M NaOH to which an additional 10 ml of 2% EDTA-0.75 M NaOH solution

was added. This solution was then passed through a cation exchange column containing 20 ml of BioRex-40 cation exchanger (20-40 mesh) preconditioned with 100 ml of 3 M HCl, 150 ml of 5% NaCl solution, and 50 ml of distilled water. The ion-exchange column was allowed to drain, washed with 60-70 ml of distilled water followed by 160 ml of 0.75 M HCl to remove any potassium (K) and/or rubidium (Rb). Cs was eluted from the column using 125 ml of 3 M HCl and the eluant evaporated to dryness on a hotplate. The residue was dissolved in 2-4 ml of water containing 1-2 drops of 8 M HNO₃, and the solution carefully transferred to a clean 50 ml C-tube using an additional 4-6 ml of distilled water. A 1 ml aliquot of 10 M NaOH was then added to the C-tube, the solution chilled in an ice-bath and Cs precipitated by drop wise addition of 2 ml of 0.12 M chloroplatinic acid (H₂PtCl₆). The precipitate (Cs₂PtCl₆) was collected on a pre-weighed GF/A filter (Whatman, 25 mm), rinsed with 2-3 ml of water, followed by 2-3 ml of acetone, and dried under an I.R. heat lamp to constant weight (±0.01 mg). The sample filters along with associated blanks and standards were all mounted on a standard ring/disc assembly, covered with a mylar film, and counted on a low background gas proportional beta counter (G5000 alpha/beta counter, Gamma Products, Inc.). The Minimum Detectable Activity (MDA) for this technique, as determined on a series of 12 reagent blank analyses, was approximately 0.01 Bq. Calibrations were performed using a standard ¹³⁷Cs solution that was indirectly traceable to the United States National Institute of Standards and Technology (NIST). The total uncertainty of the measurements was propagated from the sample counting error (taken from the standard deviation between five consecutive sample counts), the background counting error, and estimates of uncertainties associated with instrument calibration and gravimetric determination of the chemical recovery. Overall chemical recoveries varied between 48 and 87%.

Determination of ^{210}Po

A 1-10 g aliquot of dried sample material was spiked with ^{209}Po tracer and 10 mg of stable Pb carrier, and dissolved in a mixture of $\text{HNO}_3\text{-HClO}_4$ under reflux. The sample residue was converted to a chloride form by repeated evaporation with 6 M HCl, and finally dissolved in 35-40 ml of 0.5 M HCl using gentle heat. The solution was heated to 75-85°C, a few hundred milligrams of ascorbic acid added to reduce any iron (Fe) present, and Po spontaneously deposited onto the surface of a spinning silver (Ag) disc (Hamilton & Smith, 1986). ^{210}Po and ^{209}Po were measured by alpha-spectrometry using surface barrier detectors. The total uncertainty of the measurements was propagated from the sample and background counting errors, and uncertainty in calibration and addition of the ^{209}Po tracer spike.

Results

Morphological Data

Tissue samples were obtained from 3 male and 2 female adult Pacific walrus, and 1 male and 1 female bearded seal (Table 1). A recently born calf accompanied both female walrus. Available reports of walrus histological samples revealed no clinically significant lesions (animals G96-0005, G96-006, and G96-0009, T. Lipscomb, per comm., 1997). Based on physical examination and age-length relationships (Burns, 1967) both the seals were young adults. More exact determination of seal age based on layers in the teeth and claw awaits further analysis. A calf accompanied the female seal. While a visual examination indicated no obvious adverse health conditions, the seals were at the lower bound of average blubber thickness for the late spring period (after Burns and Frost, 1979). Histological material from the female found commonly occurring nematode *Parafilaroides* sp. present in the lungs, mild subacute hepatitis, and hepatic capsular fibrosis; none of which were considered to have seriously comprised

the health of the animal (T. Lipscomb, per comm., 1997).

^{137}Cs and ^{210}Po Activity Concentrations in Tissues of Walrus and Seal

The activity concentration of ^{137}Cs and ^{210}Po in muscle, liver and kidney of Pacific walrus and bearded seal are shown in Table 2a&b. Decay corrections for ^{210}Po were based on a supported ^{210}Pb activity concentration of 1 Bq kg⁻¹ (dry wt.). The majority of samples were processed within about 120 days of collection. The dry tissue weights (after lyophilization) to wet tissue weight ratios have been used to calculate radionuclide concentrations and results reported on a wet weight basis (Bq kg⁻¹, wet wt.). The mean activity concentrations of ^{137}Cs in tissues of Pacific walrus were 0.07 Bq kg⁻¹ in muscle, 0.09 Bq kg⁻¹ in liver, and 0.07 Bq kg⁻¹ in kidney (Table 2a). Similarly, mean ^{137}Cs activity concentrations in tissues of bearded seal were 0.17 Bq kg⁻¹ in muscle, 0.10 Bq kg⁻¹ in liver, and 0.18 Bq kg⁻¹ in kidney. The activity concentrations of ^{210}Po in tissues of Pacific walrus and bearded seal are considerably higher and span a much broader range compared with levels of ^{137}Cs (Table 2b). The mean (and range) of ^{210}Po concentrations in the muscle tissue, liver and kidney of Pacific walrus were 28.7 Bq kg⁻¹ (12 to 58 Bq kg⁻¹), 189 Bq kg⁻¹ (70 to 428 Bq kg⁻¹), and 174 Bq kg⁻¹ (101 to 276 Bq kg⁻¹), respectively. The mean (and range) of ^{210}Po concentration in muscle tissue, liver and kidney of bearded seal were 27 Bq kg⁻¹ (25.5-28.7 Bq kg⁻¹), 207 Bq kg⁻¹ (185-228 Bq kg⁻¹) and 68 Bq kg⁻¹ (23-113 Bq kg⁻¹), respectively.

Discussion

There has been widespread concern over the possible health and environmental impacts of radioactive waste dumped in the Kara and Barents Seas and/or held in wastewater storage ponds and reservoirs in the West Siberian Basin (Yablokov et al., 1993;

Strand & Cooke, 1995; Layton *et al.*, 1997a). The ^{137}Cs concentration in tissues of Pacific walrus and bearded seal from the Bering Sea are generally lower than previously reported concentrations in the muscle tissue of mammals from other regions (Roos *et al.*, 1992; Dahlgaard, per. comm., 1996). Roos *et al.* (1992) published some comparable baseline data for different species of seal collected from Antarctic waters but these data are typically reported as 'not detected' or with large uncertainties. The measured ^{137}Cs concentration in muscle tissue from a single sample of crabeater seal was $2.5 \pm 0.8 \text{ Bq kg}^{-1}$, dry weight. This compares with a mean average dry weight ^{137}Cs concentration in Bearded Seal collected from the Bering Sea of $0.63 \pm 0.03 \text{ Bq kg}^{-1}$ (recalculated from Table 2A). The Risø National Laboratory in Denmark has been measuring ^{137}Cs concentrations in mammals (largely whale and seal) from Greenland since the early 1960s (Dahlgaard, per. comm., 1996). During the period between 1982 and 1991, the ^{137}Cs concentration in muscle tissue averaged 0.4 Bq kg^{-1} , wet weight (N=19). The combined average ^{137}Cs concentration in muscle tissue of mammals collected from the Bering Sea during 1996 was 0.1 Bq kg^{-1} (wet wt., N=7) or a factor of 2-6 times lower concentration. Similarly, grey seal (*Halichoerus grypus*) collected from the North Sea and north-east Atlantic Ocean during 1987 appear to have contained much higher levels of ^{137}Cs with mean concentrations in muscle and liver ranging between 14.3 and 27.5 Bq kg^{-1} , and 6.4 and 14.6 Bq kg^{-1} , respectively (Anderson *et al.*, 1990). These findings were not unexpected because of localized inputs of radiocesium into the marine environment from transport of radioactive waste from European reprocessing facilities, riverine discharges into the Arctic Seas from large Siberian rivers, dumping of radioactive waste and inputs from the Chernobyl accident (Hamilton, 2004).

The concentration of ^{137}Cs in muscle tissue, kidney, and to a lesser extent, in liver of bearded seal appear to be lower than those concentrations measured in comparison tissues of Pacific walrus. The mean ^{137}Cs concentration in the

muscle tissue of bearded seal ($0.17 \pm 0.03 \text{ Bq kg}^{-1}$, wet wt.) was more than double that observed in Pacific walrus ($0.07 \pm 0.02 \text{ Bq kg}^{-1}$, wet wt.). The observed differences in radionuclide concentrations may be related to the small sample size, the age or sex of the animals, different rates of deposition and elimination from tissues, food habits, and/or different seasonal patterns of distribution. We assume that walrus and bearded seal acquire the subject elements through ingestion of prey. While bearded seal and Pacific walrus feed predominately on benthic invertebrates and there is a strong dietary overlap (in particular for the clam, *Serripes greonlandicis*) bearded seals are euryphagous and opportunistic feeders while walrus are stenophagous and specialized for infaunal feeding. Therefore, dietary habits coupled with differences in seasonal distributions of walrus and seal in the Bering and Chukchi Seas may well lead to real differences in ^{137}Cs body burdens between the two species.

In general, the activity concentrations of naturally occurring ^{210}Po in walrus and seal are between 2 and 4 orders of magnitude higher than those of ^{137}Cs . The range of ^{210}Po concentrations observed between species was also much greater than observed for ^{137}Cs . The mean dry weight ^{210}Po concentration in the muscle tissue of bearded seal is about $96 \pm 6 \text{ Bq kg}^{-1}$ (N=2) (recalculated from Table 2b). This compares with ^{210}Po concentrations reported by Roos *et al.* (1992) of 5 ± 0.3 , 4.1 ± 0.6 and $16 \pm 8 \text{ Bq kg}^{-1}$ dry weight in the muscle tissue of leopard seal, crabeater seal and Weddel seal, respectively, from the Antarctica. Based on this small population size, bearded seal from the Bering Sea appear to contain significantly higher levels of ^{210}Po compared with other species of seal from the Antarctic.

The enrichment of ^{210}Po in marine species over that of other radionuclides including the precursor ^{210}Pb has been well documented (Carvalho, 1988; Cherry and Shannon, 1974). Noshkin *et al.* (1994), Pentreath *et al.* (1979) and others have also noted similar large variations in ^{210}Po concentrations among different species and across the same species.

These differences are probably related to the feeding habits of marine species and the variable ^{210}Po content of any food consumed but, in general, the mechanisms are poorly understood. ^{210}Po has a relatively short half-life (138 days) and any seasonal variations in ^{210}Po uptake by mammals (e.g., through ingestion of different prey or prey containing different concentrations of ^{210}Po) should show in the level of ^{210}Po in tissue samples. Of those pinnipeds living in the Bering and Chukchi Sea, only the bearded seal and Pacific walrus feed predominately on benthic invertebrates (Lowly *et al.*, 1980). It is not unreasonable to assume that ^{210}Po body burdens may be very different to those of piscivorous-feeding and/or mixed-feeding mammals.

An interesting observation is that the highest ^{210}Po concentrations in our samples appear to be associated with females of both species (Table 1 & 2B; specimens G96-0009, G96-0748, and G96-0002). The female walrus we sampled were younger than the males, and, while the total number of samples analyzed is small, these results appear to differ from those of other workers reporting bioaccumulation of trace elements with age in walrus and other Arctic pinnipeds (Warburton & Seagars, 1993; Wagemann & Stewart, 1994; Seagars *et al.*, 1994; Mackey *et al.*, 1996;) and transfer of various contaminants from female pinnipeds to their pups during lactation (Wagemann *et al.*, 1988; Addison & Brodie, 1987; Anderson *et al.*, 1990) or trans-placental transmission to the fetus (Eisler, 1988). In the absence of other data, it is possible that the higher accumulation of ^{210}Po in female walrus results from undetermined sex related or species-specific factors. Diet, age and sex are known to influence the concentration of organochlorines in marine mammals (Addison and Brodie, 1987) and may be equally important for radioactive contaminants. However, a more plausible explanation might well be related to the summer-fall distributions of these groups into the northern regions of the Chukchi Sea and

regional differences in the distribution of ^{210}Po .

Field-Derived Concentration Factors

The bioconcentration factors for ^{137}Cs and ^{210}Po in Pacific walrus and bearded seal from the Bering Sea are shown in Table 3. Field-derived CF values have been expressed in $\text{m}^3 \text{kg}^{-1}$, and as a unitless measure to help facilitate comparisons with data from the literature. CF values for ^{137}Cs and ^{210}Po were estimated using associated seawater concentrations of 2 and 1 Bq m^{-3} , respectively (Povinec *et al.*, 1996; Ellis *et al.*, 1995).

The full range of CF values (unitless) for ^{137}Cs are as follows: 24-47 for muscle tissue, 41-49 for liver, and 28-44 for kidney in Pacific walrus, and 71-100 for muscle tissue, 47-57 for liver, and 85-90 for kidney in bearded seal. The mean CF values for muscle tissue in walrus and seal are 36 and 86, respectively. Our data for the muscle tissue of marine mammals from the Bering Sea are in general agreement with a world-wide consensus value of 100 reported by Povinec *et al.* (1996) and compares with CF values of 100 for fish flesh, and 30 for molluscs and crustacea, as previously recommended by IAEA (IAEA, 1985).

For ^{210}Po , CF values for each tissue type are very similar across the two species (Table 3). The mean ^{210}Po CF values (unitless) for muscle, liver and kidney tissues were 29,000, 190,000 and 174,000, in Pacific Walrus, and 27,000, 206,000 and 208,000, in bearded seal, respectively. Recommended CF values for different groups of marine organisms in the Arctic Seas were recently reported by Templeton *et al.*, (1997) for use in risk assessments. Based on limited data from Roos *et al.* (1992), the recommended CF values for mammals are given as 1,700 for muscle tissue and 22,000 for liver. Measured ^{210}Po CF values for Pacific walrus and bearded seal from the Bering Sea are up to an order of magnitude higher for each of these two tissue types.

Exposure to Radionuclides in Subsistence Diets

In evaluating doses to individuals exposed to radionuclides in subsistence diets, it is important to have detailed information on dietary patterns of subject populations as well as accurate radiometric data. Detailed information related to the subsistence diets of Alaskan Natives and/or Alaskan coastal communities were not available for this assessment. However our studies clearly show that the concentration of ^{137}Cs in tissues of Pacific walrus and bearded seal are several orders of magnitude lower than those of naturally occurring ^{210}Po . Existing background concentrations in waters of the Chukchi Sea are in order of 2 Bq m^{-3} (Ellis *et al.*, 1995). The highest predicted ^{137}Cs concentration in seawater resulting from an instantaneous release from nuclear wastes in the Russian Arctic is estimated to be around 0.03 Bq m^{-3} (Layton *et al.*, 1997b) or well below existing background levels. As such, the potential risks associated with ingestion of foods derived from walrus and seal containing ^{137}Cs derived from Russian nuclear wastes will be extremely low, and pose no threat to human health. Based on the activity concentrations of ^{137}Cs and ^{210}Po measured in seal and walrus from the Bering Sea, and using dose conversion factors adopted from the International Commission of Radiological Protection (ICRP) for ingestion of radionuclides, i.e., $1.2 \times 10^{-8} \text{ Sv Bq}^{-1}$ for ^{137}Cs (ICRP 30, 1978) and $1.2 \times 10^{-6} \text{ Sv Bq}^{-1}$ for ^{210}Po (ICRP 67, 1994), the committed effective dose for adults from intakes of ^{137}Cs will be insignificant or about 3-4 orders of magnitude lower compared with that from naturally-occurring ^{210}Po . In evaluating these data, it is important to recognize that unsupported ^{210}Po has a relatively short half-life ($T_{1/2} = 138$ days), and a decay factor between catch and consumption should be used. More extensive studies will also be needed to evaluate the relatively high ^{210}Po CF values in mammals from the Bering Sea compared with values reported from other regions. Furthermore, polar bear are known to track directly across the Bering Sea from the Asian continent during winter providing an important

source of meat for coastal communities. It would be useful to supplement the data presented here with a more detailed study of the levels and transfer pathways of radionuclides in Alaskan arctic regions, e.g., through consumption of polar bear, whale, and caribou meat. These data could be used to develop a more comprehensive assessment of background radiation doses for critical population groups, and yield more accurate dose estimates for any potential releases and/or transfers of radionuclides into the region.

Conclusion

The focus of this study was to provide high quality baseline data on radionuclide concentrations in marine mammals as a basis for conducting an integrated risk assessment on Alaskan coastal communities exposed to radionuclides in subsistence diets. The activity concentration of ^{137}Cs and naturally occurring ^{210}Po were measured in the muscle tissue, kidney and liver of Pacific walrus and bearded seal collected from the Bering Sea. The mean ^{137}Cs concentrations (wet weight) in tissues of Pacific walrus and bearded seal were 0.07 and 0.17 Bq kg^{-1} in muscle, 0.09 and 0.10 Bq kg^{-1} in liver, and 0.07 and 0.175 Bq kg^{-1} in kidney, respectively. Bioconcentration factors for muscle and kidney tissues in Pacific walrus appear to be significantly lower than for bearded seal, and values for both species are generally lower than those reported for other pinniped species. Observed differences in the concentration of ^{137}Cs in muscle and kidney tissue between the two species can be explained by differences in prey utilized by these two benthic feeding mammals, and in turn, may be coupled to differences in their seasonal distribution within the Bering and Chukchi Seas. The activity concentrations and associated bioconcentration factors for naturally-occurring ^{210}Po are much higher than those of ^{137}Cs . Consequently, the committed effective dose from ingestion of ^{137}Cs in foods derived from walrus and seal will be insignificant or about 3-4 orders of magnitude lower

compared with that from ^{210}Po . Using a worst-case release scenario, we also conclude that any potential risks associated with consumption of marine foods containing ^{137}Cs derived from the dumping of nuclear wastes will be extremely low and pose no threat to human health. There is, however, a general lack of measurement data and information on the levels and transfer behaviors of radionuclides through Arctic food chains leading to man. A key to more accurate dose estimates will be the availability of high quality radiometric data covering all potential exposure pathways, and detailed dietary information for critical population groups.

Acknowledgement

This study formed part of a contribution to the Arctic Nuclear Waste Assessment Program (ANWAP) and was made possible through the cooperation of the U.S. Fish and Wildlife Service (Marine Mammals Management Field Office-AK). We gratefully acknowledge the assistance of boat captains Mr. Tason Nowpowkahok, Mr. Dennis James, Sr., H.V. Slwooko, Jr., Mr. David Angie, and L. Apangoqlook for sample collection. We also thank Marshall Stuart for sample preparation; Jennifer Luna, Patricia Lopez, and Rayla Bradsher for secretarial support, and Lynn Wilder for preparing graphics. Work performed under the auspices of the U.S. Department of Energy by the University of California at the Lawrence Livermore National Laboratory under contract W-7405-Eng-48.

References

Addison, R.F., & Brodie, P.F. 1987. Transfer of organochlorine residues from blubber through the circulatory system to milk in the lactating grey seal, *Halichoerus grypus*. *Canadian Journal of Fisheries and Aquatic Science* **44**, 782-786.

Anderson, S.S., Livens, F.R., & Singleton, D.L. 1990. Radionuclides in grey seals. *Marine Pollution Bulletin* **21**, 343-345.

Antonelis, G.A., Melin, S.R., & Bukhtiyarov, Y.A. 1994. Early spring feeding habits of bearded seals (*Erignathus barbatus*) in the Bering Sea. *Arctic* **47**, 74-79.

Arctic Monitoring and Assessment Programme (AMAP) (1998a), AMAP Assessment Report: Arctic Pollution Issues, P. Strand et al., (editors), Oslo, Norway, xii + 859 pp.

Arctic Monitoring and Assessment Programme (AMAP) (1998b), AMAP Assessment Report: Arctic Pollution Issues, Chapter 8: Radioactivity, P. Strand et al., (editors), Oslo, Norway, xii + 525-619.

Bengston, J.L., & Sniff, D.B. 1981. Reproductive aspects of female seals (*Lobodon carcinophagus*) along the Antarctic Peninsula, *Canadian Journal of Zoology* **59**, 92-102.

Bradley, D.J., & Jenquin, U.P. 1995. Radioactive Inventories and Sources for Contamination of the Kara Sea by Riverine Transport, Pacific Northwest National Laboratory, Richland, WA. PNNL Report PNWD-2316, 1995.

Braham, H.W., Burns, J.J., Fedoseev, G.A., & Krogman, B.D. 1984. Habitat partitioning by ice-associated pinnipeds: distribution and density of seals and walruses in the Bering Sea, April 1976. In: Fay F.H., and Fedoseev G.A., editors. Soviet-American Cooperative Research on Marine Mammals, Vol. 1- Pinnipeds, pp. 25-47, NOAA Technical Report NMFS-12, 104 pp.

Burns, J.J. 1967. The Pacific bearded seal, Federal Aid in Wildlife Restoration Project Reports, Volume VIE, Alaska Department of Fish and Game, Juneau, AK. Project W-6-R and W-14-R, 66 pp.

- Burns, J.J., & Frost, K.J. 1979. Natural history and ecology of the bearded seal, *Erignathus barbatus*, Outer Continental Shelf Environmental Assessment Program (OCSEAP), Boulder, CO, Final Report RU 230, 77 pp.
- Carvalho, F. P. 1988. ^{210}Po in marine organisms: A wide range of natural radiation dose domains. *Radiation Protection Dosimetry* **24**, 113-117.
- Cherry, R.D., & Shannon, L.V. 1974. The alpha radioactivity of marine organisms, *Atomic Energy Review*. **12**, 47-65.
- Dahlgaard, H. 1996. Personal Communication.
- Eisler, R. 1988. Arsenic hazards to fish, wildlife, and invertebrates: a synoptic review, U.S. Fish and Wildlife Service Biological Report. **85**(1.12), 92 pp.
- Ellis, K.M., Smith, J.N., Nelson R.P., Kilius, L., MacDonald, R., Carmack, E., & Moran, S.B. 1995. Distribution of Artificial Radionuclides in the Arctic Ocean from 1994. In: Proceedings. Second International Conference on Environmental Radioactivity in the Arctic, Strand P. & Cooke A., editors, Østerås, Norway, 204-297.
- Fay, F.H., Kelly B.P., & Sease J.L. 1986. Determination of age of walrus taken in recent spring harvests in the Bering Strait region. Fish and Wildlife Service, Anchorage, AK. Final Report No. WR-0439-85, U.S., 110 p.
- Fay, F.H., Bukhtiyarov, Y.A., Stoker, S.W., & Shults, L.M. 1984. Foods of the Pacific walrus in winter and spring in the Bering Sea. In: Fay F.H. and Fedoseev G.A., editors. Soviet-American Cooperative Research on Marine Mammals, Vol. 1-Pennipeds, pp. 81-88. NOAA Technical Report NMFS-12, 104 pp.
- Fay, F.H. 1982. Ecology and biology of the Pacific Walrus, *Odobenus rosmarus divergens* Illiger., U.S. Fish & Wildlife Service, North American Fauna No.74, Washington, DC, 279 pp.
- Fay, F.H. & Stoker S.W. 1982a. Analysis of reproductive organs and stomach contents from walrus taken in the Alaskan Native harvest, Spring 1980. Final Report, Contract 14-16-007-81-5216 to U.S. Fish and Wildlife Service, Anchorage, AK, 86 pp.
- Fay, F.H. & Stoker S.W. 1982b. Reproductive success and feeding habits of walrus taken in the 1982 spring harvest, with comparisons from previous years. Final Report, Eskimo Walrus Commission, Nome, AK, 91 pp.
- Fay, F.H., Feder, H.M., & Stoker S.W. 1977. An estimate of the impact of the Pacific Walrus population on its food resources in the Bering Sea. U.S. Department of Commerce, National Technical Information Service (Springfield, VA) PB-273-505, 38 pp.
- Fay, F.H. 1974. The role of ice in the ecology of marine mammals of the Bering Sea. In: Oceanography of the Bering Sea, Hood D.W., and Kelley E.K., editors, Institute of Marine Science, University of Alaska, Fairbanks, 383-399.
- Finely, K.J., & Evans, C.R. 1983. Summer diet of the bearded seal (*Erignathus barbatus*) in the Canadian high Arctic. *Arctic* **36**, 82-89.
- Fedoseev, G.A. 1990. The role of ice in changes of Pacific Walrus distribution and numbers. In: The Ecology and Management of Walrus Populations, Fay F.H., Kelly B.P., & Fay B.A., editors—Report of an International Workshop, pp 185-186. Final Report Marine Mammal Commission, Contract T68108850, NTIS PB91-100479, 186 pp.

- Gilbert, J.R., Fedoseev, G.A., Seagars, D.J., Razlivalov, E., & Lachugin, A. 1992. Aerial census of Pacific Walrus, 1990. U.S. Fish and Wildlife Service, Administrative Report R7/MMM 92-1, Anchorage, AK, 33 pp.
- Hamilton T.F. 2004. Linking legacies of the cold war to arrival of anthropogenic radionuclides in the oceans through the 20th century. In: Marine Radioactivity, Livingston H., editor, Elsevier Applied Science Publication, Chapter 2, Lawrence Livermore National Laboratory, CA, UCRL-JC-201419, 65 pp.
- Hamilton, T.F., Millies-Lacroix, J-C, & Hong, G.H. 1996. ^{137}Cs (^{90}Sr) and Pu isotopes in the Pacific Ocean: Sources & Trends. In: Radionuclides in the Oceans: Inputs and Inventories, Guéguéniat P, Germain P, and Métivier H, editors, Institut de Protection et de Sureté Nucleire, France, Les Éditions de Physique, Les Ulis cedex A, France, pp 29-58.
- Hamilton T.F. & Smith J.D. 1986. Improved alpha-energy resolution for the determination of polonium isotopes by alpha-spectrometry. *Applied Radiation and Isotopes* **37**, 628-30.
- International Atomic Energy Agency (IAEA). 1985. Sediment Kds and Concentration Factors for Radionuclides in the Marine Environment, Technical Reports Series No. 247, IAEA, Vienna, 73 pp.
- International Commission on Radiation Protection. 1978. Limits for Intakes of Radionuclides by Workers, Annuals of the ICRP, ICRP Publication 30, Pergamon Press, Oxford, U.K, 116 pp.
- International Commission on Radiation Protection. 1994. Age-dependent doses to members of the public from intakes of radionuclides: Part 2, Ingestion dose coefficients. Annuals of the ICRP, ICRP Publication 67, Vol. 23, No. 3/4, Pergamon Press, Oxford, U.K.
- Laws R.M. 1962. Age determination of pinnipeds with special reference to growth layers in the teeth, *Z. Saugetierkd.* **27**,129-146.
- Layton D., Vaughn P., Yarrington L., Maxwell R., Preller R., Bean J., Fuhrmann M., Rigor I., Tucker W., & Carrol J. 1997a. Transport of radionuclides in the Arctic Seas. In: Radionuclides in the Arctic Seas from the Former Soviet Union: Potential Health and Ecological Risks. Layton D., Edson L.C.D.R.R, Varela M., and Napier B., editors, Arctic Nuclear Waste Assessment Program (ANWAP), Office of Naval Research (ONR), pp. 3-1/3-35.
- Layton D., Vaughn P., Yarrington L., Maxwell R., Preller R., Bean J., Carroll J., Fuhrmann M., Rigor I., & Tucker W. 1997b. Transport of radionuclides in the Arctic Seas. In: Radionuclides in the Arctic Seas from the Former Soviet Union: Potential Health and Ecological Risk. Layton D, Edson L.C.D.R.R, Varela M, and Napier B., editors, Arctic Nuclear Waste Assessment Program (ANWAP), Office of Naval Research (ONR), Chapter 3, pp 3/1-3/36.
- Lipscomb T.L. 1997. Personal Communication.
- Lowry L.F., Frost K.J., & Burns JJ. 1980. Feeding of bearded seals in the Bering and Chukchi Seas and trophic interaction with Pacific walruses. *Arctic* **33**, 330-342.
- Lowry L.F., Frost J.K., & Burns J.J. 1979. Trophic relationships among ice inhabiting Phocid seals and functionally related marine mammals, Final report of Beaufort Sea activities, OCSEAP contract 03-5-022-53, 55 pp.
- Mackey E.A., Becker P.R., Demiralp R., Greenburg R.R., Koster B.J., & Wise S.A. 1996. Bioaccumulation of vanadium and other trace metals in livers of Alaskan cetaceans and pinnipeds. *Archives of Environmental Contamination and Toxicology* **30**, 503-512.

- McLaren I.A. 1958. Some aspects of growth and reproduction of bearded seal, *Erignathus barbatus* (Erxleber), *Journal Fisheries Research. Board of Canada* **15**, 219-227.
- Noshkin V.E., Robison W., & Wong K.M. 1994. Concentration of ^{210}Po and ^{210}Pb in the diet at the Marshall Islands. *Science Total Environment* **155**, 87-104.
- Pentreath R.J. & Alington D.J. 1988. Dose to man from the consumption of marine seafoods: A comparison of the naturally occurring ^{210}Po with artificially produced radionuclides, Proceedings of the International Radiation Protection Association **7**, 82-1885.
- Pentreath R.J., Lovett M.B., Harvey B.R., & Ibbett R.D. 1979. Alpha-emitting nuclides in commercial fish species caught in the vicinity of Windscale, United Kingdom, and their radiological significance to man, In: IAEA Symposium on Biological Implications of Radionuclides Released from Nuclear Industries, Vol. II, IAEA-SM-237/1, IAEA, Vienna, pp. 227-245.
- Povinec P.P., Baxter M.S., Fowler S.W., Boisson F., Carrol J., Gayol J., Liong We Kwong L., Miquel J-C., & Osvath O. 1996. Final Report on the IAEA International Arctic Sea Assessment Program, International Atomic Energy Agency, Vienna, Austria.
- Rissanen K., Ikäheimonen T.K., Matishov D., & Matishov G.G. 1997. Radioactivity levels in fish, benthic fauna, seals and sea birds collected in the northeast Arctic of Russia. In: Radionuclides in the Oceans RADOCS 96-97, Radioprotection—colloques. Germain P., Guary JC, Guéguénat P, and Métivier H, editors, Vol. 32, C2, Les Éditions de Physique, Les Ulis cedex A, France, pp. C2-323/C2-331.
- Roos P., Holm E., Persson R.B.R., & Neilsen S.P. 1992. Flux of ^{210}Pb , ^{137}Cs , $^{239,240}\text{Pu}$, ^{238}Pu , and ^{241}Am . 1992. The Antarctic Peninsula Area. Presented at the meeting of Nordic Liason Committee for Nuclear Science.
- Scheffer V.B. 1967. Standard measurements of seals. *Journal of Mammalogy* **48**, 459-262.
- Seagars D.J., Messing P., Bowlby C.E., Noonan L., & Burns D.M. 1995. Annual Summary: Information Collected during the 1993 Spring Walrus Harvest in Alaska, U.S. Fish and Wildlife Service, Technical Report MMM 95-1, 31 p.
- Seagars D.J., Bowlby C.E., Burn D., & Garlich-Miller J. 1994. Metal concentrations in livers and kidney tissues of Pacific walrus: 1991. *Zaslouovo* Bering Sea research cruise samples. In: Abstracts of the 45th Arctic Science Conference, Anchorage, AK and Vladivostok, Russia, 11 pp.
- Strand P. & Cooke A. 1995 (editors). Environmental Radioactivity in the Arctic. Proceedings of an International Conference on Environment Radioactivity in the Arctic, Østerås, Norway, 427 pp.
- Templeton W., Harrison F., Knezovich J., Fisher N., & Layton D. 1997. Bioconcentration of Radionuclides in Marine Food-Web Organisms. In: Radionuclides in the Arctic Seas from the Former Soviet Union: Potential Health and Ecological Risks, Layton D, Edson LCDRR, Varela M, and Napier B., editors, Arctic Nuclear Waste Assessment Program (ANWAP), Office of Naval Research (ONR), pp. 4-1/4-12.

Yablokov A.V., Karasev V.K., Rummyantsev V.M., Kokeyev M. Ye., Petrov O.I., Lysyov V.N., Yemelyanenko A.F., & Rubtsov P.M.. 1993. Facts and Problems Related to Radioactive Waste Disposal in Sea Adjacent to the Territory of the Russian Federation. Moscow, Office of the President of the Russian Federation, 72 p.

Wagemann R, & Stewart REA. 1994. Concentrations of heavy metals and selenium in tissues and some foods of walrus (Obodenus rosmarus rosmarus) from the eastern Canadian Arctic and sub-arctic, and associations between metals, age and gender. *Canadian Journal of Fisheries and Aquatic Science* **51**, 426-436.

Wagemann R., Stewart R.E.A., Lockhart W.L., & Stewart B.E. 1988. Trace metals and methyl mercury: associations and transfer in harp seal (Phoca groenlandica) mothers and their pup, *Marine Mammal Science* **4**, 339-355.

Warburton J., & Seagars D J. 1993. Heavy metal concentrations in liver and kidney tissues of Pacific walrus. U.S. Fish and Wildlife Service Technical Report, MMM 94-1, 23pp.

Table 1. Sample collection data for Pacific Walrus and Bearded Seal from the Bering Sea

USFWS field log #	Collection date	Species type	Animal physiology
G96-0003	10-May-96	Pacific Walrus	Adult male
G96-0005	11-May-96	Pacific Walrus	Adult male, 322 cm in length
G96-0006	14-May-96	Pacific Walrus	Adult male, 305 cm in length
G96-0009	19-May-96	Pacific Walrus	Young adult female, 290 cm in length
G96-0748	20-May-96	Pacific Walrus	Young adult female, 270 cm in length
G96-9001	12-May-96	Bearded Seal	Adult male, 215 cm in length
G96-9002	17-May-96	Bearded Seal	Adult female, 202 cm in length

Table 2. Concentration of ^{137}Cs and ^{210}Po in walrus and bearded seal from the Bering Sea

USFWS field log #	Muscle Tissue			Liver			Kidney		
	dry wt. (%)	Activity Concentration (Bq kg ⁻¹ , wet wt.)		dry wt. (%)	Activity Concentration (Bq kg ⁻¹ , wet wt.)		dry wt. (%)	Activity Concentration (Bq kg ⁻¹ , wet wt.)	
		¹³⁷ Cs	²¹⁰ Po		¹³⁷ Cs	²¹⁰ Po		¹³⁷ Cs	²¹⁰ Po
Pacific Walrus (<i>Odobenus rosmarus divergens</i>)									
G96-0003	27.3	0.071±0.004	20.3±0.9	30.9	0.094±0.006	89±2	21.5	0.057±0.003	123±7
G96-0005	29	0.047±0.003	12.6±0.7	27.9	0.084±0.003	70±4	21.2	0.068±0.005	101±4
G96-0006	26.7	0.066±0.004	27.7±1.0	27.7	0.098±0.004	87±5	21.2	0.067±0.005	112±6
G96-0009	29.9	0.080±0.003	25.3±0.9	28.9	0.089±0.003	273±16	24.3	0.088±0.005	257±15
G96-0748	29.5	0.095±0.004	57.6±2.0	29.2	0.082±0.003	428±26	26.7	0.057±0.003	276±15
Bearded Seal (<i>Erignathus barbatus</i>)									
G96-9001	29.1	0.14±0.01	25.5±1.0	27.8	0.093±0.004	228±13	22.7	0.17±0.01	113±7
G96-9002	31.6	0.20±0.01	28.7±1.1	30.9	0.113±0.004	185±10	22.6	0.18±0.01	142±9

Notes: ^{210}Po was decay corrected to the date of sampling assuming a supported ^{210}Pb concentration of 1 Bq kg⁻¹ (dry wt.).

Table 3. Bioconcentration factors for ^{137}Cs and ^{210}Po in Pacific Walrus and Bearded Seal from the Bering Sea

Radionuclide	Species	Tissue type	Tissue concentration, Bq kg ⁻¹ (dry wt.)	Bioconcentration factor		
				Range	Mean	Unitless
^{137}Cs	Pacific Walrus (<i>Odobenus rosmarus divergens</i>) (N=5)					
		muscle	0.05-0.1	0.024-0.047	0.036	36
		liver	0.08-0.1	0.041-0.049	0.045	45
		kidney	0.06-0.09	0.028-0.044	0.034	34
		Bearded Seal (<i>Erignathus barbatus</i>) (N=2)				
		muscle	0.14-0.20	0.071-0.10	0.086	86
^{210}Po	Pacific Walrus (<i>Odobenus rosmarus divergens</i>) (N=5)					
		muscle	13-58	13-58	29	29000
		liver	70-428	70-428	190	190000
		kidney	101-276	101-276	174	174000
		Bearded Seal (<i>Erignathus barbatus</i>) (N=2)				
		muscle	25-29	25-29	27	27000
	liver	185-228	185-228	206	206000	
	kidney	113-142	113-142	208	208000	

Notes: Bioconcentration factors for ^{137}Cs and ^{210}Po have been calculated using associated seawater concentrations of 2 and 1 Bq m⁻³, respectively.

Sample Locations

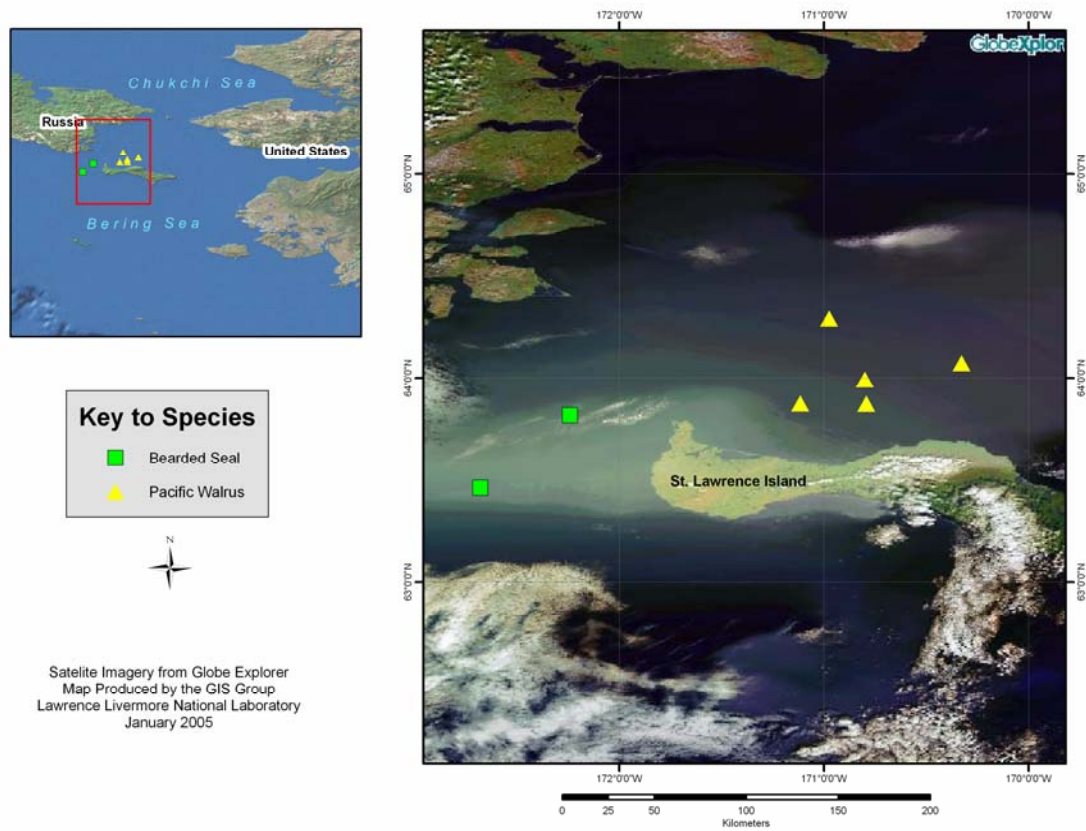


Fig. 1. Map showing sample locations off St. Lawrence Island in the Bering Sea.