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# Fukushima derived radiocesium in subsistence-consumed northern fur seal and wild celery

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ABSTRACT

In July 2014, our investigative team traveled to St. Paul Island, Alaska to measure concentrations of radiocesium in wild-caught food products, primarily northern fur seal (*Callorhinus ursinus*). The 2011 Fukushima Daiichi Nuclear Power Plant accident released radiocesium into the atmosphere and into the western Pacific Ocean; other investigators have detected Fukushima-derived radionuclides in a variety of marine products harvested off the western coast of North America. We tested two subsistence-consumed food products from St. Paul Island, Alaska for Fukushima-derived radionuclides: 54 northern fur seal, and nine putchki (wild celery, *Angelica lucida*) plants. Individual northern fur seal samples were below minimum detectable activity concentrations of $^{137}$Cs and $^{134}$Cs, but when composited, northern fur seal tissues tested positive for trace quantities of both isotopes. Radiocesium was detected at an activity concentration of 37.2 mBq $^{134}$Cs kg$^{-1}$ f.w. (95% CI: 35.9–38.5) and 141.2 mBq $^{137}$Cs kg$^{-1}$ f.w. (95% CI: 135.5–146.8). The measured isotopic ratio, decay-corrected to the date of harvest, was 0.26 (95% CI: 0.25–0.28). The Fukushima nuclear accident released $^{134}$Cs and $^{137}$Cs in roughly equal quantities, but by the date of harvest in July 2014, this ratio was 0.2774, indicating that this population of seals has been exposed to small quantities of Fukushima-derived radiocesium. Activity concentrations of both $^{134}$Cs and $^{137}$Cs in putchki were below detection limits, even for composited samples. Northern fur seal is known to migrate between coastal Alaska and Japan and the trace $^{134}$Cs in northern fur seal tissue suggests that the population under study had been minimally exposed Fukushima-derived radionuclides. Despite this inference, the radionuclide quantities detected are small and no impact is expected as a result of the measured radiation exposure, either in northern fur seal or human populations consuming this species.

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## 1. Introduction

The 2011 Tohoku earthquake triggered powerful tsunami waves up to 40 m high that devastated coastal Japan more than 5 km inland (Mori et al., 2011). Nearly 16,000 people died as a result of the earthquake and tsunami, and an additional 3000 people are still listed as missing (Mimura et al., 2011). In the wake of this devastation, the Fukushima nuclear power plant (NPP) lost electric power and the ability to circulate coolant, resulting in overheating of its nuclear reactors. A series of overpressure ventings, combined with the buildup of explosive hydrogen, ultimately led to loss of containment and releases of radionuclides to the atmosphere and the marine environment. While the atmospheric release has been abated, marine releases may be ongoing due to groundwater seepage (WNA, 2015).

The atmospheric plume from the Fukushima Daiichi reactors traveled east and passed over North America days after the initial release. Modeling (Behrens et al., 2012; Rossi et al., 2013), has predicted that the leading edge of the marine plume reached the west coast of North America sometime in 2014. Measurements (Smith et al., 2015) indicate that Fukushima-derived radionuclides

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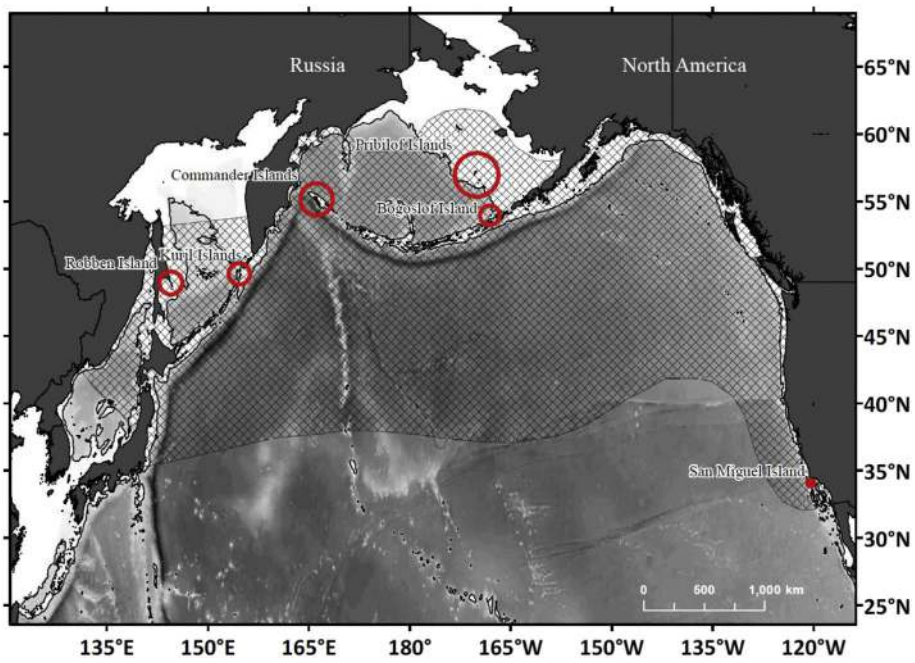


**Fig. 1.** Range of northern fur seal, with the relative abundance of the breeding areas represented by the relative size of red circles. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

were present in waters above the Canadian continental shelf by June 2013, with concentrations off coastal North America expected to increase through 2015 before declining. Due to Pacific Ocean currents, the Fukushima plume likely arrived on the west coast of North America prior to transport north and east toward our study site (Smith et al., 2015).

The arrival of the Fukushima marine plume has aroused concern for some North American stakeholders, particularly those living near the coast and those who consume seafood from the Pacific Ocean. Additionally, the potential effects of exposure to Fukushima-derived radionuclides on sensitive marine species will need to be studied and understood in the coming years. To date, investigators have primarily focused on exposure in marine fish (Johansen et al., 2015; Madigan et al., 2012; Neville et al., 2014). To complement existing work, this study measured activity concentrations of Fukushima-derived radionuclides in northern fur seal (*Callorhinus ursinus*), both to understand the radiological risk to the animals, as well as the risk associated with human consumption of animal tissues.

Northern fur seal is an eared seal that is widely distributed throughout the North Pacific Ocean and Bering Sea (Fig. 1). These animals spend the majority of their time foraging at sea, returning to land on six island groups across the Bering Sea and North Pacific Ocean (National Marine Fisheries Service, 2007). Individuals may travel great distances, particularly during their seasonal migration. Adult females and juvenile male fur seal from the Pribilof Islands annually migrate from the Bering Sea to the Central North Pacific Ocean, ranging as far west as the Okhotsk, Kurile, and the Japanese Current, returning each spring to the Pribilof Islands (Ream et al., 2005).

Radiocesium biomagnifies through marine foodwebs (Calmet et al., 1991; Carroll et al., 2002; Heldal et al., 2003; Kasamatsu and Ishikawa, 1998; Watson et al., 1999). Thus the northern fur seal, a predator, should be an excellent sentinel of marine radiocesium in the North Pacific. Additionally, due to this biomagnification effect, human exposure to radiocesium via consumption of fur seal meat is of concern. The approximately five hundred Aleuts on the Pribilof Islands of St. Paul and St. George have annually harvested between 323 and 608 (mean = 328 individuals harvested) two-to-four year old male fur seal for subsistence food in the last decade (Zimmerman and Letcher, 1986; Zimmerman and Melividov, 1987). Edible meat per animal ranges from 10.4 to 12.5 kg, with a mean of 11.9 kg animal$^{-1}$, for a mean total of 3903 kg harvested annually (8.1 kg per person) (Zimmerman and Letcher, 1986; Zimmerman and Melividov, 1987). Finally, the species has been designated as "depleted" under the Marine Mammal Protection Act of 1986 due to the decline of the population by more than 50% since the 1950s (NMFS, 2007). Several stakeholder and government groups are interested in demonstrating protection of the public and of northern fur seal populations exposed to Fukushima-derived radionuclides, and so testing of northern fur seal tissues for radiocesium is warranted.

## 2. Material and methods

Tissue samples were collected over a two-week period during the subsistence harvest season on St. Paul Island in July 2014. Northern fur seal samples were collected during the community harvest on July 24, 2014 (MMPA permit 14327) from 54 individual sub-adult males. Approximately 500 g of skeletal muscle was collected from the neck of each animal and refrigerated until dehydration. Additional biological samples included nine putchki plants (wild celery, *Angelica lucida*), which is the dominant ground cover on St. Paul Island and is occasionally consumed by residents. Putchki samples were collected from several sites over the harvest period; only the edible stalks of the plant were processed.

All biological samples were field-dried at 60 °C to achieve a stable mass using a commercial food dehydrator. Dehydration occurred within 18–24 h and the stabilized samples were subsequently transported to Colorado State University (CSU), where they were dry-ashed in a muffle furnace at 450 °C for 4–6 h. CSU's muffle furnace does not allow the user to set a temperature ramp, but the furnace generally reached full temperature after about two hours. Ashes were hand-homogenized and counted on a high

purity germanium (HPGe) detector for 23 h. The counting procedure utilized a constant geometry so that sample results would be inter-comparable. The HPGe detector (ORTEC Model GMX-80230-S, S/N 31-N40266A) has a crystal that is 76.4 mm diameter by 79.5 mm long, with a 0.76 mm Be window, a relative efficiency of 95% with a 2.3 keV resolution at 1.33 MeV. Spectral analysis was performed using Canberra Lynx digital signal processor and Genie 2000 software (Canberra Industries, Inc. Meridian, CT). Radiocesium concentration in samples was quantified using the $^{137}$Cs 662 keV photopeak, and the $^{134}$Cs 605 keV photopeak. Fig. 2 presents the energy calibration curve for CSU's gamma detector system. Minimum detectable activities, calculated using the Currie equation (Currie, 1968), were 95.6 mBq $^{134}$Cs and 84.2 mBq $^{137}$Cs, corresponding to tissue activity concentrations of 192 mBq $^{134}$Cs kg$^{-1}$ w.m. and 168 mBq $^{137}$Cs kg$^{-1}$ w.m., for a theoretical 500 g w.m. sample.

There is no commercially available radionuclide standard for marine mammal tissue, so the method described above was calibrated against the International Atomic Energy Agency's fish flesh standard (IAEA, 2004) and losses were propagated throughout the data. Table 1 displays certified radionuclide concentrations in IAEA-414, and Fig. 3 depicts the acquired gamma spectrum.

**Table 1**
Certified radionuclide values for IAEA-414 as of January 1, 1997.

| Radionuclide | Certified value | Unit | 95% Confidence interval |
|---|---|---|---|
| $^{40}$K | 481 | Bq/kg | 470 – 486 |
| $^{137}$Cs | 5.18 | Bq/kg | 5.12 – 5.22 |
| $^{232}$Th | 0.028 | Bq/kg | 0.025 – 0.031 |
| $^{234}$U | 1.22 | Bq/kg | 1.15 – 1.26 |
| $^{235}$U | 0.050 | Bq/kg | 0.045 – 0.055 |
| $^{238}$U | 1.11 | Bq/kg | 1.07 – 1.15 |
| $^{238}$Pu | 0.0230 | Bq/kg | 0.0221 – 0.0250 |
| $^{239+240}$Pu | 0.120 | Bq/kg | 0.116 – 0.123 |
| $^{241}$Am | 0.197 | Bq/kg | 0.193 – 0.204 |

## 3. Results and discussion

### 3.1. Tissue activity concentrations of radiocesium in northern fur seal

Activity concentrations of radiocesium in individual northern fur seal samples were below minimum detectable quantities. To obtain an average estimate of the radiocesium content by increasing the total quantity of radiocesium being analyzed, samples were combined electronically by summing the individual spectra. When all northern fur seal samples were combined, both $^{137}$Cs and $^{134}$Cs were detectable; Fig. 4 shows the composite gamma spectrum. Table 2 depicts the major energies used for gamma analysis, while Table 3 shows the results of spectrum deconvolution. Separate minimum detectable activities were calculated for the composite spectrum: 684 mBq $^{134}$Cs and 602 mBq $^{137}$Cs, corresponding to tissue activity concentrations of 26.3 mBq $^{134}$Cs kg$^{-1}$ w.m. and 23.2 mBq $^{137}$Cs kg$^{-1}$ w.m. The activity concentration of radiocesium (decay corrected to July 25, 2014, the date of tissue harvest) in the composted seal tissue was 37.2 mBq $^{134}$Cs kg$^{-1}$ w.m. (95% CI: 35.9–38.5) and 141.2 mBq $^{137}$Cs kg$^{-1}$ w.m. (95% CI: 135.5–146.8). These activity concentrations are miniscule compared to the American food standard for radiocesium of 1200 Bq kg$^{-1}$ ($^{137}$Cs + $^{134}$Cs) as well as the more restrictive Japanese standard of 100 Bq kg$^{-1}$. The isotopic ratio of $^{134}$Cs/$^{137}$Cs in northern fur seal was 0.26 (95% CI: 0.24–0.28). The Fukushima releases produced $^{134}$Cs and $^{137}$Cs in roughly equal amounts (Chaisan et al., 2013). Assuming an equal quantity and then correcting for decay, the ratio on the date of harvest was 0.2774, a ratio that agrees well with the value measured in this study. Table 4 compares the results

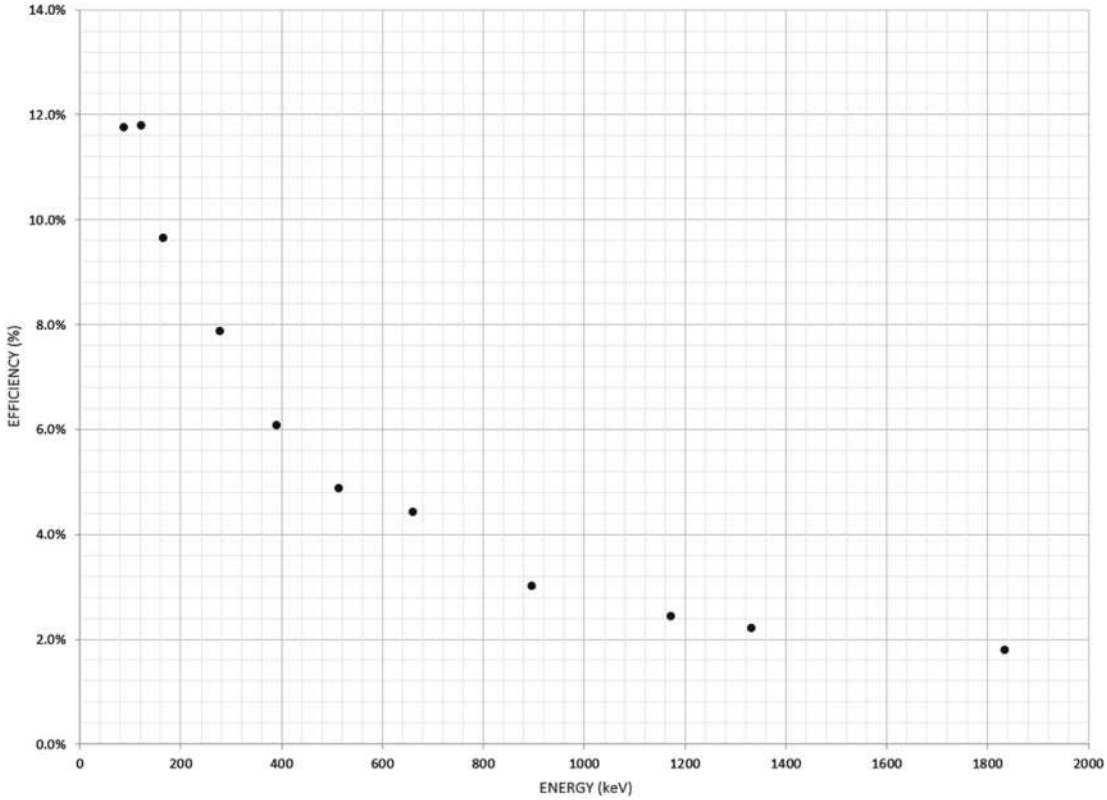


**Fig. 2.** Energy calibration curve for Colorado State University's HPGe detector system.

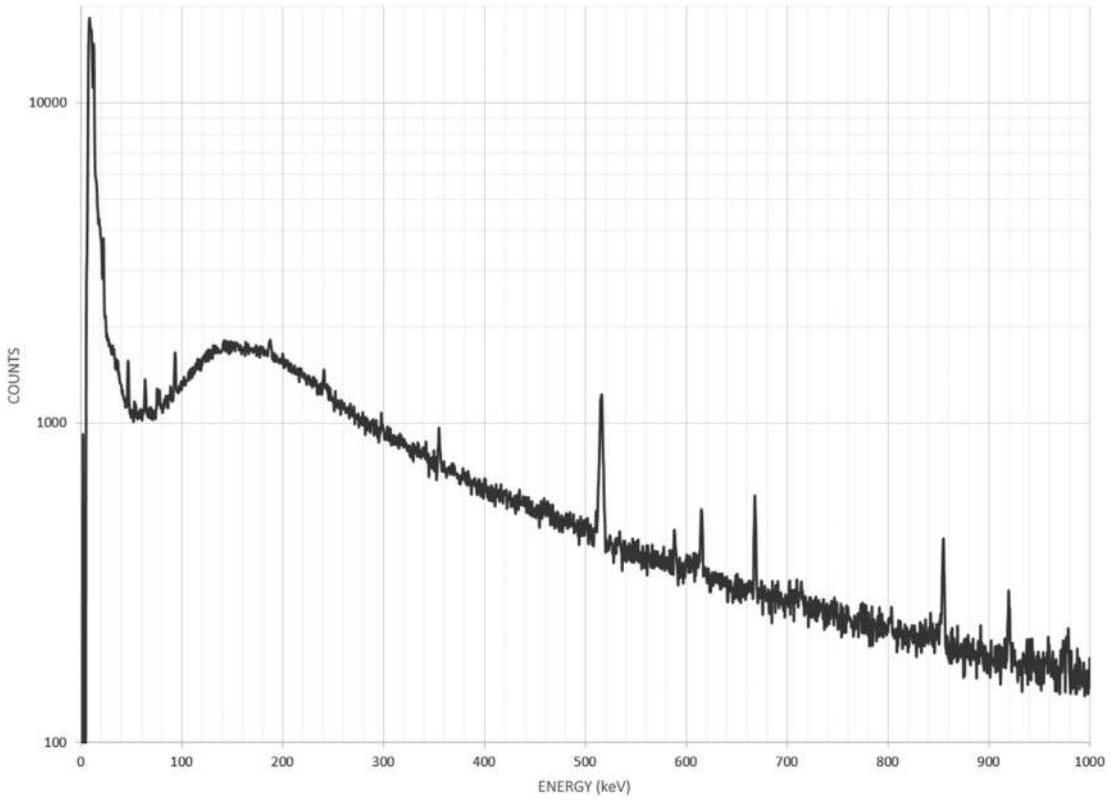


**Fig. 3.** Gamma spectrum of IAEA-414 fish flesh standard. Note 662 keV $^{137}$Cs peak.

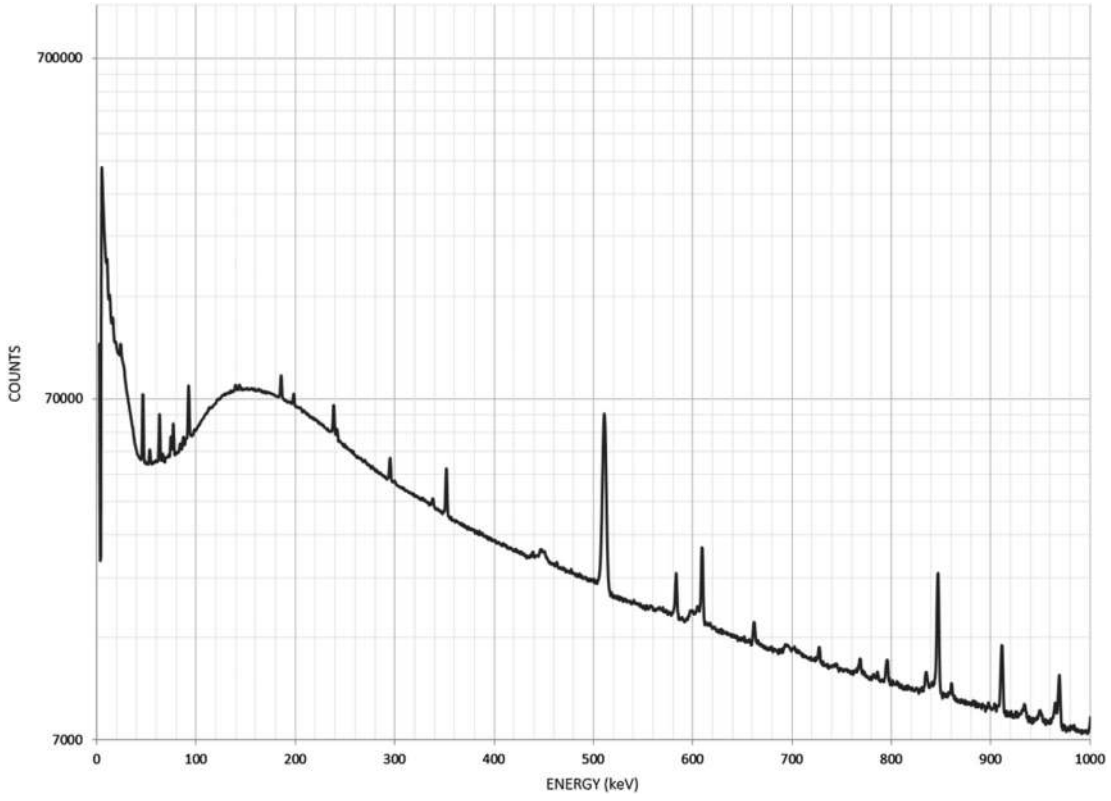


**Fig. 4.** Composited gamma spectrum of 54 individual northern fur seals showing 605 and 795 keV $^{134}$Cs peak and 662 keV $^{137}$Cs peak. $^{228}$Ac also contributes to 795 keV peak.

**Table 2**
Major energies (<1000 keV, >1% Iγ) of isotopes contributing to the measurement of $^{134}$Cs and $^{137}$Cs. Underlined values are those peaks used to determine activity concentration. Star (*) indicates peaks used to de-convolute the spectrum.

| Isotope | Energy, Iγ |
|---|---|
| $^{134}$Cs | 475 keV, 1.5% |
| | 563 keV, 8.4% |
| | 569 keV, 15.4% |
| | 605 keV, 97.6% |
| | 795 keV, 85.5% |
| | 801 keV, 8.7% |
| $^{137}$Cs | 662 keV, 85.1% |
| $^{214}$Bi | 609 keV, 46.1% |
| | 665 keV, 1.5% |
| | 769 keV, 4.9% (*) |
| | 806 keV, 1.2% |
| | 934 keV, 3.0% |

**Table 3**
Deconvolution of the composited spectrum. 665 keV $^{214}$Bi net counts estimate was obtained by adjusting the 769 keV $^{214}$Bi peak net counts for energy efficiency and Iγ.

| Peak energy (keV) | Net counts ± 1σ | Contributing isotopes |
|---|---|---|
| 605 | 2251 ± 47 | $^{134}$Cs |
| 769 | 2791 ± 52 | $^{214}$Bi |
| 662 | 10828 ± 104 | $^{214}$Bi + $^{137}$Cs |
| 665 | 959 ± 18[a] | $^{214}$Bi |
| 662 | 9869 ± 105[a] | $^{137}$Cs |

[a] Calculated value.

of this study to previous studies of radiocesium in pinnipeds, mostly from the North Atlantic. Northern fur seal exposure to Fukushima radionuclides likely occurred via the consumption of fish or other prey from areas of the Pacific Ocean contaminated by the Fukushima marine release.

Brown et al. (2006) created a probabilistic biokinetic model of radiocesium uptake in ringed (*Phoca hispida*) and other seals exposed, via the food chain, to a constant water concentration of 2mBq L$^{-1}$ of $^{137}$Cs. Brown uses the model to predict tissue activity concentrations of radiocesium in seal populations given a known quantity of radiocesium in the water column. We opted to use this model to do the opposite: to predict the maximum concentration of radiocesium in water to which the seals under study were exposed, based upon our measurements of radiocesium in seal tissue. This quantity gives some information about the migration patterns of the northern fur seal population from which our individuals were harvested.

Northern fur seal undertake an approximately 240-day winter migration (Ream et al., 2005) from the Western Pacific to the Pribilof Islands. During this migration period they would be exposed to ever-diminishing quantities of radiocesium. As such, it is not possible to establish an exact source term for radiocesium exposure along the migration: the seals' route is not understood sufficiently well to draw conclusions about when or whether the seals crossed through the highest concentration areas of Fukushima marine plume. Despite this, it is still possible to calculate a theoretical maximum concentration of radiocesium to which seals could be exposed by assuming that the majority of exposure occurred in the Western Pacific, and only negligible exposure occurred during the migration period. Brown et al. (2006) allometrically derived a depuration rate for $^{137}$Cs for a 50 kg ringed seal of 0.01 d$^{-1}$, and proposes a range of 0.0063–0.0148 d$^{-1}$ based upon the work of others (ICRP, 1979; Whicker and Schultz, 1982). A sub-adult male fur seal may weigh 30 kg; using the same allometric approach with a lower mass yields a depuration rate very similar to Brown's. From this rate of 0.01 d$^{-1}$, we estimate that 91% of a sub-adult male fur seal's radiocesium body burden is eliminated over the course of an approximately 240-day winter migration (Ream et al., 2005) from the Western Pacific to the Pribilof Islands. This corresponds to an estimated tissue concentrations of 0.41 Bq $^{134}$Cs kg$^{-1}$ w.m. and 1.6 Bq $^{137}$Cs kg$^{-1}$ w.m at the start of the migratory period. In turn, these tissue concentrations correspond to estimated concentrations of 1.5 mBq $^{134}$Cs L$^{1}$ and 5.8 mBq $^{137}$Cs L$^{-1}$ in the water column. These estimated concentrations are several orders of magnitude lower than measurements (Caffrey et al., 2012) and modeled estimates (Behrens et al., 2012) of radiocesium in seawater off coastal Japan immediately following the Fukushima releases, and are comparable to more recent measurements (Smith et al., 2015) of radiocesium in the Eastern Pacific. Thus, while this northern fur seal population

**Table 4**
The result of this study compared with previously reported values of radiocesium in pinnipeds. Reported concentrations are for muscle tissue.

| Year | Species | Location | Tissue concentration | Reference |
|---|---|---|---|---|
| 2014 | *C. ursinus* | St. Paul Island, USA | 37.2 mBq $^{134}$Cs kg$^{-1}$ w.m.<br>141.2 mBq $^{137}$Cs kg$^{-1}$ w.m. | This study. |
| 1968 | *P. groenlandicus* | Magdalen Islands, Canada | 133 Bq $^{137}$Cs kg$^{-1}$ ash (juvenile)<br>85.1 Bq $^{137}$Cs kg$^{-1}$ ash (adult) | (Samuels et al., 1970) |
| 1980 | Unknown seal | Barents Sea | 1.1 Bq $^{137}$Cs kg$^{-1}$ d.w. | (Holm et al., 1983) |
| 1984 | Unknown seal | Greenland | 0.53 Bq $^{137}$Cs kg$^{-1}$ [a]<br>0.79 Bq $^{137}$Cs kg$^{-1}$ [a] | (Aarkrog et al., 1985) |
| 1984 | *C. ursinus* | Sanriku, Japan<br>Bering Sea | 0.25–0.37 Bq $^{137}$Cs kg$^{-1}$ [a] | (Nagaya and Nakamura, 1987) |
| 1987 | Unknown seal | Greenland | 0.078 Bq $^{137}$Cs kg$^{-1}$ [a]<br>0.25 Bq $^{137}$Cs kg$^{-1}$ [a] | (Aarkrog et al., 1989) |
| 1987 | *H. grypus* | North Rona, UK<br>Isle of May, UK | 6.4–27.5 Bq $^{137}$Cs kg$^{-1}$ [a] | (Anderson et al., 1990) |
| 1988 | Unknown seal | Greenland | 0.24 Bq $^{137}$Cs kg$^{-1}$ [a] | (Aarkrog et al., 1992) |
| 1988–1995 | *P. vitulina* | Ireland & UK | undetectable | (Watson et al., 1999) |
| 1988–1995 | *H. grypus* | Ireland & UK | undetectable | (Hamilton et al., 2008; Watson et al., 1999) |
| 1996 | *E. barbatus* | St. Lawrence Island, USA | 0.14 Bq $^{137}$Cs kg$^{-1}$ f.w.<br>0.20 Bq $^{137}$Cs kg$^{-1}$ f.w. | (Hamilton et al., 2008) |
| 2003 | *P. hispida* | Spitsbergen, Greenland | 0.40–0.61 Bq $^{137}$Cs kg$^{-1}$ f.w. | (Andersen et al., 2006) |
| 2002 | *C. cristata* | North Greenland Sea | <0.2–0.42 Bq $^{137}$Cs kg$^{-1}$ f.w. | (Andersen et al., 2006) |
| 2000–2002 | *E. barbatus* | Spitsbergen, Greenland | 0.06–0.42 Bq $^{137}$Cs kg$^{-1}$ f.w. | (Andersen et al., 2006) |
| 2003 | *P. groenlandicus* | South Barents Sea | <0.2–0.53 Bq $^{137}$Cs kg$^{-1}$ f.w. | (Andersen et al., 2006) |

[a] Fresh or dry weight basis not reported.

has been exposed to small quantities of Fukushima-derived radiocesium, the population likely has not been exposed to the higher concentrations found within the main body of the Fukushima marine plume.

### 3.2. Tissue activity concentrations of radiocesium in putchki

The underlying pathway of radiocesium exposure is very different for terrestrial plants compared to marine mammals. Putchki and other terrestrial plants could only have been exposed to radiocesium via atmospheric deposition from the Fukushima releases onto St. Paul Island. Given the large distance between St. Paul Island and coastal Japan (c.a., 4000 km), as well as the time since the release, it would be reasonable to expect only very low concentrations of radiocesium in putchki and other terrestrial plants. However, the underlying geology of St. Paul Island is volcanic rock. Where topsoil exists, it is thin and relatively nutrient-poor. Under these conditions, it might be easy to draw a parallel to the well-known case of Bikini Atoll's coconut palms (Robison and Stone, 2002), which, due to potassium deficiency, preferentially absorbed radiocesium following radionuclide deposition from nuclear weapons tests. Contrary to this reasoning, a study conducted in Iceland (Sigurgeirsson et al., 2005) reported that radiocesium binds strongly to volcanic soils, preventing transport through the soil column and inhibiting uptake into vegetation. Thus, we expect that terrestrial samples from St. Paul Island would contain only the smallest quantities of Fukushima-derived radiocesium. True to this prediction, levels of radiocesium were below detectable activity concentrations in putchki, even when samples were combined.

## 4. Conclusions

Trace quantities of Fukushima-derived radionuclides ($^{134}$Cs and $^{137}$Cs) were detected in the muscle tissue of the northern fur seal, but not in native plants (putchki) harvested from St. Paul Island in July, 2014. Northern fur seal is a marine predator that migrates annually to the Pacific Ocean and as such is likely to bioaccumulate radiocesium, making it an effective sentinel species. Now that the plume is dispersed across a broader area of the Pacific Ocean, the younger seals consumed for subsistence purposes in future years are unlikely to bioaccumulate greater quantities than those seals that spent their first few winters foraging within the most concentrated plume and were harvested and sampled in 2014. We only detected radiocesium in combined tissue samples, and well below activity concentrations of naturally occurring radionuclides such as $^{4}$K. Radiocesium is unlikely to cause health impacts in northern fur seal or the human populations consuming this species.

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