

Measurements of cesium in Arctic beluga and caribou before and after the Fukushima accident of 2011



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ABSTRACT

Concern from northern communities following the Fukushima Daiichi nuclear accident of March 2011 has prompted a reassessment of the safety of their traditional foods with respect to radioactivity levels. To this end, a study was conducted to measure the levels of radionuclides in Arctic caribou (*Rangifer tarandus*) and beluga (*Delphinapterus leucas*). The main radionuclide of concern is cesium-137, which is easily transferred through the lichen-caribou food chain. Previous studies have been conducted on the cesium-137 levels in Canadian caribou herds from 1958 to 2000, allowing researchers to determine the amount of cesium-137 in caribou specifically attributable to atmospheric weapons testing and the Chernobyl nuclear accident in 1986. In this study, samples of lichens, mushrooms, caribou, beluga and beluga prey collected before and after the Fukushima accident were analyzed for radioactivity levels. Samples were processed and measured using gamma ray spectroscopy to identify the radionuclides present and determine the radioactivity concentration. Both calibration standards and Monte Carlo simulations were used to determine the efficiency of the detectors for the samples, taking into account differences in individual sample sizes as well as matrices. In particular, a careful analysis of the atomic composition of lichens and mushrooms was performed to ensure the efficiencies for these sample types were correct. A comparison of the concentrations from before and after the accident indicated that there was no increase in radioactivity as a result of the atmospheric plume from the Fukushima accident. Some cesium-137, likely attributable to fallout from atmospheric weapons testing of the 1950s and 1960s (since there was no cesium-134 measured in the samples), was measured in the post Fukushima caribou and beluga whale samples; however, this amount was determined to be insignificant for any radiological concern (9.1 ± 1.8 and 0.63 ± 0.23 Bq kg⁻¹ ww respectively). The activity concentrations of cesium-137 was about 200 times smaller than that of natural radioactive potassium in the beluga samples. Both the caribou and beluga results showed that these foods continue to be a healthy food choice for northern Canadians with respect to radioactivity, and this result has been communicated to the nearby northern communities and stakeholders.

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1. Introduction

On March 11, 2011, the largest ever recorded earthquake in Japan (magnitude 9 on the Richter scale) occurred. The epicentre was slightly more than 180 km away from the Fukushima Daiichi nuclear power plant, which had 6 reactors.

Details of the accident can be found in UNSCEAR (2014) and details about the Health Canada radiation monitoring effort with respect to the accident can be found in Health Canada (2015).

The main radionuclide of concern was cesium-137 (¹³⁷Cs), which has a half-life of 30 years and is a surrogate for potassium in biological systems, easily accumulating in plants and animals. ¹³⁷Cs has been a health concern since the 1950s when significant amounts were released during nuclear weapons testing. Nuclear accidents, such as Chernobyl in 1986 and Fukushima-Daiichi in 2011, have also

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been sources of ^{137}Cs in the environment. Cesium-134 (^{134}Cs), which is also released into the environment from nuclear accidents but has a relatively short half-life (~2 years) is a useful indicator of recent inputs of Cs into the environment. Because the measurements in this study were performed within a reasonable time (well within 5 half-lives) after the Fukushima accident, it is possible to attribute ^{137}Cs to the accident based on the presences of a proportional quantity of ^{134}Cs . Conversely, the absence of ^{134}Cs in the samples indicates that ^{137}Cs in samples is the result of earlier environmental releases.

The amount of ^{137}Cs released to the atmosphere between March 12, 2011 and April 6, 2011, is estimated to be 9.66 PBq or approximately 20% of the release from Chernobyl. The estimated source term also includes 11.3 PBq of ^{134}Cs (WHO, 2012). According to recent estimates (Health Canada, 2015), relatively little contamination was deposited in the Canadian Arctic (1–10 Bq m⁻² for ^{137}Cs).

Following the Fukushima Daiichi nuclear accident in March 2011, northern Canadians expressed concern about the levels of radioactive contaminants in important traditional foods. Therefore, a study was conducted to measure the levels of radionuclides in Arctic caribou (*Rangifer tarandus*) and beluga whale (*Delphinapterus leucas*).

1.1. Other sources of radiocesium in the environment

1.1.1. Radiocesium from nuclear weapons fallout

According to UNSCEAR, weapons testing contributed a total 960 PBq of ^{137}Cs into the atmosphere (Annex E, Section 2, subsection J of UNSCEAR, 1982). Atmospheric nuclear weapons testing started in 1945 and, for the most part, ended in 1963 with the signing of the Partial Test Ban Treaty, at which time the US and the Soviet Union stopped atmospheric tests. Despite the limited nuclear weapons testing since the signing of the Partial Test Ban Treaty, the largest input of radiocesium to the environment occurred in the 1950s and 1960s. Approximately 1 EBq (1000 PBq) of ^{137}Cs was released to the biosphere during this time, with 90% of it being produced by atmospheric testing (NCRP 154, 2007; UNSCEAR, 1982).

1.1.2. The Chernobyl accident (1986)

In addition to the fallout from atmospheric nuclear tests, fallout from the nuclear accident in Chernobyl on April 26, 1986 also reached the Canadian Arctic. The amount of ^{137}Cs released from the Chernobyl accident was determined to be 54 PBq according to the 1995 estimates in Eisenbud and Gesell (1997). This amount of ^{137}Cs is about 6% of that released to the environment by all atmospheric weapons tests combined. While the accident also expelled other radioisotopes into the atmosphere, most of these additional isotopes were short lived and were undetectable after a few months. After the Chernobyl accident, increased levels of ^{137}Cs and ^{134}Cs were found in caribou, lichens, and reindeer throughout the northern hemisphere (Åhman and Åhman (1994), Aarkrog et al. (2000), Strandberg (1997), Pålsson et al. (1994), Taylor et al. (1988), Macdonald et al. (2007)). ^{134}Cs was measured in muscle samples of Canadian caribou herds with a mean concentration between 20 and 32 Bq/kg (Macdonald et al., 2007). This amount of ^{134}Cs was used to estimate the amount of ^{137}Cs in the caribou attributed to the Chernobyl accident. It was estimated that, on average, 20% of the ^{137}Cs in the caribou was due to Chernobyl. Mean levels of ^{134}Cs in the Porcupine herd after the Chernobyl accident from 1986 to 1990 were as high as 7.2 Bq kg⁻¹ (Macdonald et al., 2007); however, these Cs levels from Chernobyl in caribou were well within the limits of applicable standards for food (Health Canada, 2000; CODEX, 1995). The total amount of deposition in

Canada of $^{134,137}\text{Cs}$ was 415 TBq (Health and Welfare Canada, 1987), dividing this by the area of Canada 9.98×10^6 km², one estimates the average deposition in Canada as ~42 Bq m⁻² from the Chernobyl accident. Huda et al. (1988) break this mean ground deposition down to 12 Bq m⁻² of ^{134}Cs and 30 Bq m⁻² of ^{137}Cs .

1.2. Exposure pathways for radiocesium in the Arctic environment

1.2.1. Radiocesium in the caribou food chain

In the Arctic, alkali minerals such as potassium are incorporated into the development of lichen, a symbiotic organism of algae and fungi, where they bioaccumulate. Cs acts as a surrogate for potassium in the environment (Nieboer et al., 1978; Hanson 1980). More specifically, the bioaccumulation of ^{137}Cs in lichens is mainly due to the absorption by the fungal partner, in order to satisfy its potassium requirements (Hofmann et al., 1993). The natural concentration of potassium in the Arctic is low (Hanson, 1980), which may result in greater Cs bioaccumulation in Arctic lichens. A pathway for ^{137}Cs to enter the human food supply exists considering that, during the winter months, lichen is the primary food source for Arctic caribou and caribou meat is the main source of protein for some northern Canadian residents. Because of this, previous studies have been made on the ^{137}Cs levels in Canadian Arctic caribou herds from 1958 to 2000. By using the ratio of ^{137}Cs to the shorter lived ^{134}Cs , Macdonald et al. (2007) were able to determine the amount of ^{137}Cs in caribou that was specifically attributable to the Chernobyl accident and to atmospheric nuclear weapons testing.

In northern ecosystems, biological processes are slow, as exemplified by low turnover rates for vegetation (litter-fall as a proportion to standing crop). This is due to a short growing season and a limited supply of nutrients, moisture and heat energy. Therefore in Arctic environments, man-made contaminants generally endure the dissipation and degradation processes longer than in temperate regions (Taylor et al., 1985). In contrast, in southern and temperate zones biological systems have effectively “cleansed” themselves of fallout radioisotopes, through fast biomass throughput and a removal of surface fallout by precipitation into various sinks (Taylor et al., 1985). Although in temperate regions the total amount of fallout remains the same after correcting for radioactive decay, the fallout becomes dissipated to lower soil strata or deposited in sediments of lakes and rivers. These locations tend to be out of the reach of the recycling process of the ecosystem (Svoboda and Taylor, 1979). The fact that radionuclide contaminants are more likely to remain accessible in the Arctic ecosystem than in a temperate ecosystem is one of the reasons why the assessment of the caribou food chain is important in this unique environment.

1.2.2. Radiocesium in the beluga food chain

Generally, the marine ecosystem concentration of ^{137}Cs is lower than in the terrestrial ecosystem (Strand et al., 1998; Avery, 1996). Few studies have been conducted on the measurements of Cs in marine mammals. In these studies, the highest concentration of Cs in marine mammals was measured off the U.K. coast, followed by Lake Baikal (seals); concentrations tend to decrease toward southern sampling points (Yoshitome et al., 2003). Marine mammals, especially beluga whales are an important food source for Inuit (Usher, 2002; Harwood and Smith, 2002). Following the Fukushima accident, specific concerns were raised by Inuvialuit (Inuit from the western arctic) hunters who harvest beluga whales from the Eastern Beaufort Sea stock. This stock of whales winters in the Bering Sea and forms large aggregations in the Mackenzie Estuary of the Beaufort Sea in the summer (Richard et al., 2001; Hauser et al., 2014). To date there are no known published reports on radionuclide levels in beluga whales circumpolarly.

There are two ways to model radioactivity in the marine food web, the concentration factor approach (this is the steady state approach); and the dynamic food chain and uptake model which targets tissues and organs and takes into account competition between ions in marine water (Rudie Heling, private communication). For the purpose of this work, we will be considering the concentration factor approach.

2. Material and methods

2.1. Samples

Porcupine caribou muscle samples were obtained from an ongoing contaminant monitoring program based in Old Crow, Yukon. Lichens (*Cladina rangiferina* and *Flavocetraria cucullata*) and mushrooms (*Russola* sp. and *Lactarius* sp.) were collected from the Porcupine caribou wintering area near Chapman Lake in central Yukon. Pre Fukushima samples were archived at $-50\text{ }^{\circ}\text{C}$ until analyzed for this study. Collection dates are specified in Table 1. Note that no pre Fukushima lichen samples were collected.

The beluga and potential beluga prey samples were obtained for analysis of both source and exposure to the predator beluga. Beluga samples were obtained from the Hendrickson Island beluga monitoring program (see Fig. 1). While belugas from the Eastern Beaufort Stock summer aggregate throughout the Mackenzie Estuary (Harwood et al., 2014), the samples for the study were specifically obtained from the Hendrickson Island monitoring program. This stock winters in the Bering Sea from December to April, where they are more likely to be affected by the radioactive plume from the Fukushima-Daiichi site that is being dispersed in the Pacific Ocean. A fist-sized sample of muscle was taken from the dorsal region and was frozen at $-20\text{ }^{\circ}\text{C}$ until further analysis. The details of the sampling procedure can be found in Loseto et al. (2015).

The beluga prey samples were obtained from a fish sampling program at Shingle Point in Shallow Bay within the Mackenzie Estuary (just northwest of Inuvik in Fig. 1) where beluga aggregate for the summer (Harwood et al., 2014). Fish species were selected to cover a range of feeding and life histories that may provide insight into both variation among species and exposure to belugas. The beluga prey samples included arctic cisco (*Coregonus autumnalis*), inconnu (*Stenodus leucichthys*), lake whitefish (*Coregonus clupeaformis*), broad whitefish (*Coregonus nasus*), long nosed sucker (*Catostomus catostomus*), saffron cod (*Eleginus gracilis*), least cisco (*Coregonus sardinella*), and starry flounder (*Platichthys stellatus*). It should be noted that samples of Arctic cod (*Boreogadis saida*), which are thought to be a critical component of the beluga diet (Loseto et al., 2009), were not available.

In both cases of the caribou and the beluga samples, sampling timing and location is very dependent on the hunters, the samples were donated as part of the hunters' usual hunting pattern. The general location of these samples was two most westerly populations of these animals in Canada.

Unlike the atmospheric transport of Cs to the Arctic, which can occur over a period of a couple of weeks, the transport of

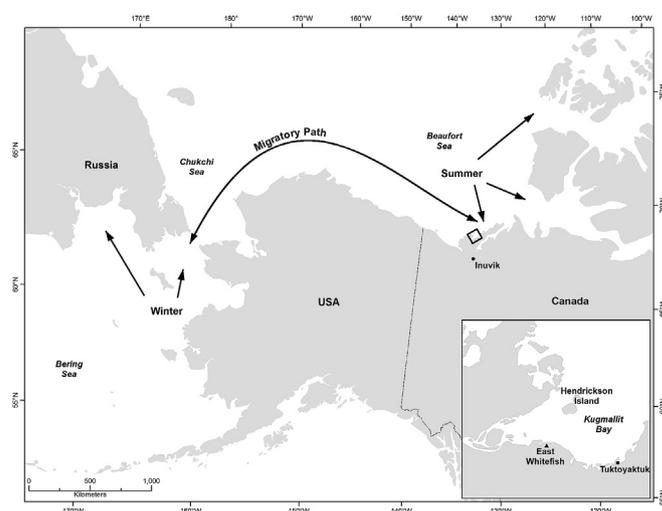


Fig. 1. The beluga migration pattern of the beluga under study and the location of the sampling program. Hendrickson Island (sampling location) is indicated in the zoomed in square which is located just north of Inuvik in the larger map.

radionuclides in the ocean is a slow process. At this point in time it is unclear when the plume of radioactivity from Fukushima will arrive in the Arctic Ocean, although there have been estimates of the ocean plume dispersion from Fukushima. Rossi et al. (2013) have estimated that only a small amount of ^{137}Cs will enter the Bering Strait into the Arctic. A recent study by Smith et al. (2015) has shown that ^{134}Cs from Fukushima had reached 1500 km west of British Columbia by June 2012. By June 2013, this signal had spread onto the Canadian continental shelf in the Pacific. That study also reported results from the CCGS Louis S. St. Laurent in the Beaufort Sea, in September 2012, which only found ^{137}Cs . These results indicate that as of September 2012, radioactivity from Fukushima had not yet reached the Arctic Ocean by ocean current transport through the Bering Sea. This is important to consider with respect to our results. The measurements on beluga and beluga prey in this paper are still valuable as a confirmatory study for the atmospheric dispersion of contamination from Fukushima and perhaps as a baseline for the Fukushima marine plume. Note that according to Buesseler (2014) the amount of ^{137}Cs deposited in the ocean from the Fukushima atmospheric plume was 10–50 PBq and from the oceanic plume it was 4–40 PBq. These two pathways are both comparable in size. Further measurements of radiocesium in beluga and beluga prey taking place after the arrival of the marine plume in the Bering Sea would be valuable.

All samples were sent to the Environment Canada Laboratory in Burlington so that they could be freeze dried and homogenized prior to measurement by γ -ray spectroscopy. Since this analysis used samples that had been collected for other studies, the volumes and masses were not consistent. To correct for this, each sample was placed in a cylindrical Parkway jar, the thickness (cylindrical height) and the density of each sample was measured and, Monte Carlo techniques were used to simulate the efficiency of the

Table 1
The sampling dates.

Sample type	Pre Fukushima date range	Post Fukushima date range
Caribou (n = 20,14)	11-Sep-2009 to 29-Nov-2009	17-Aug-2011 to 25-Sep-2011
Lichens (n = 11)	–	5-Aug-2011
Mushrooms (n = 6,6)	7-Aug-2010	5-Aug-2011
Beluga (n = 19,22)	5-Jul-2010 to 27-Jul-2010	6-Jul-2011 to 24-Jul-2011
Beluga prey (n = 15,23)	20-Jul-2010 to 27-Jul-2010	28-Jul-2012

detector for each sample matrix.

For some of the beluga samples, there were tendons which could not be homogenized. Since these tendons were a different density, making the samples heterogeneous, they were carefully removed from the samples before the remainder of the sample was measured using γ -ray spectroscopy.

2.2. Monte Carlo modelling of the detector efficiencies

Since the samples were all different masses and volumes, Monte Carlo techniques were used to calculate the High Purity Germanium (HPGe) detector efficiencies for these different sample geometries. The Monte Carlo code which was used for this purpose was EGSnrc 4r2.0.0 (Kawrakow and Rogers, 2003). To ensure that the Monte Carlo simulations were set up correctly, previously measured efficiency standards were modelled. For more information on this please see Stocki et al. (2016).

2.3. Measurement and analysis of the samples

The samples were measured using a High Purity Germanium detector (HPGe) within a Canberra Gamma Analyst system. This system has a robotic sample changer which automatically changes the samples and measures them. The efficiency of this detector at 1.33 MeV is ~48% relative to a 3" by 3" diameter NaI(Tl) detector. The detector used was a Canberra BE5030.

One of each of the caribou, beluga, and beluga prey samples was measured for 55 h; all others were measured for 12 h. The samples that were measured for longer time were chosen to be the most sensitive (in terms of higher efficiencies) out of all the different sample sizes. The lichen and mushroom samples were measured for between 12 and 24 h.

The samples were then analyzed with the Genie 2000 software (See Canberra website), and with Fitzpeaks [see <http://www.jimfitz.co.uk/>]. The calculations of the activity concentration were done in a Microsoft Excel spreadsheet to facilitate the use of the simulated efficiencies.

Because of the concern around the Fukushima accident, the samples were analyzed for ^{134}Cs and ^{137}Cs . The spectra were also searched for photopeaks of other anthropogenic radionuclides. Potassium-40 (^{40}K) was used as a control for the caribou and beluga samples to ensure the system was operating correctly, as it has been previously measured in animals at a fairly consistent level of 100 Bq kg^{-1} for wet weights (ww). The room background levels of ^{40}K were subtracted from the sample results. Other naturally occurring radionuclides were also observed in the spectra, but these were not quantified.

All of the results were decay corrected to the sampling date.

There were some lichen samples for which neither ^{137}Cs nor ^{40}K were observed, so a geometric mean was calculated by excluding the values which were less than or equal to zero. In this case, an arithmetic mean was calculated and found to be 3.9 Bq kg^{-1} ww for ^{137}Cs and 7.5 Bq kg^{-1} ww for ^{40}K .

2.4. Determination of environmental transport parameters

There are three parameters which could be calculated in terms of lichen-caribou pathway modelling. There is the aggregated transfer coefficient (T_{ag}) which is the activity concentration (Bq kg^{-1}) in the food product divided by the radionuclide deposition in soil (Bq m^{-2}) (AMAP, 2004; Palsson et al. 2009). For the calculations of T_{ag} in this study, the amount of fallout deposited on the ground was used and decay corrected. This transfer factor changes with time. Second, there is the transfer coefficient (Howard et al., 2009), which depends on how much lichen the caribou eat.

The transfer coefficient is the equilibrium ratio of the activity concentration in meat to the daily dietary radionuclide intake rate. Finally, there is also the concentration ratio (Leppänen, 2014). It is simply the ratio of the activity concentrations in the caribou (ww) to the lichen (dw) or caribou (ww) to the mushroom (dw) (Leppänen, 2014; Howard et al., 2009), where ww is wet (fresh) weight and dw is dry weight. In this paper, we have calculated all three of these parameters in order to help guide future environmental pathway modelling.

For the marine samples, bioconcentration factors (BCFs) are the ratio of the contaminant concentration in the whole body (or the tissue) of the organism of interest to the concentration in the water. These ratios can be either dimensionless or have dimensions of L kg^{-1} or $\text{m}^3 \text{ kg}^{-1}$. This is consistent with the concentration factor for marine systems defined in TRS 422 (IAEA, 2004). This parameter represents the net effects of all bioaccumulation and elimination processes affecting the transfer of a radionuclide from food, water, and particulate matter (ingested and/or filtered) to the organism (ANWAP, 1997). For the fish considered here, for Cs, the IAEA working group recommended number for the Arctic BCF is $0.1 \text{ m}^3 \text{ kg}^{-1}$, similarly for whales it is also $0.1 \text{ m}^3 \text{ kg}^{-1}$ (ANWAP, 1997).

3. Results

3.1. Caribou, mushroom and lichen results

Cesium-134 was not detected (below the detection limits as presented in Table 2) in any of the lichen, mushroom, or caribou samples taken before or after the Fukushima nuclear accident. Table 3 depicts the results from this study for the Porcupine caribou, mushroom, and lichen samples, respectively. In Table 3 the geometric means of the activity concentrations of ^{137}Cs are listed. There were some cases where the radioisotope was not detected in some samples. In these cases the geometric means of ^{137}Cs have been calculated by removing these zeros.

As further discussed in Section 4.1, the caribou results in this work are lower than the levels measured in 1990 (Macdonald et al., 2007) and agree with the effective half-life (as defined in Macdonald et al. (2007) and Taylor et al. (1985)) for this particular herd. Also the amount of ^{137}Cs in the post Fukushima caribou samples is slightly less than in the samples from before the accident. As shown in Table 3, the amount of ^{40}K in the caribou samples has a geometric mean of 80.1 Bq kg^{-1} ww before the Fukushima accident and a geometric mean of 76.4 Bq kg^{-1} ww after the accident. Both of these values are close to the 100 Bq kg^{-1} result that was identified as a check for quality control (Macdonald et al., 2007). A *t*-test was done on the natural logarithmic transformed distributions and it was found that these two distributions are not significantly different at the significance level of 0.05.

Previous work (Hofmann et al., 1993) has also showed that the amount of ^{40}K is not consistent (in lichens), and is dependent on the availability of potassium from either air or soil and from the effect of leaching by precipitation. So unlike the caribou and the beluga case where potassium is regulated within those animals, in lichens there is no consistent value of ^{40}K with which one can compare.

The geometric mean of ^{137}Cs in the mushrooms samples before and after the accident are relatively consistent at 10 Bq kg^{-1} ww and 9.1 Bq kg^{-1} respectively. Potassium-40 in these samples was 80.97 Bq kg^{-1} and 71.9 Bq kg^{-1} pre and post Fukushima respectively.

In the caribou, mushrooms, and lichen samples, there were no other unusual radionuclides (namely, anthropogenic gamma emitting radionuclides) found in the individual spectra.

Table 2
Minimum Detectable Concentrations (MDC) for the different individual samples for ^{134}Cs .

Sample type	^{134}Cs MDC (Bq kg^{-1} ww)	^{137}Cs MDC (Bq kg^{-1} ww)	Proportion of ^{137}Cs assuming ^{134}Cs at detection limit (Bq kg^{-1} ww)
Caribou	2.8	1.6	6.2
Lichen	3.8	3.9	8.4
Mushrooms	0.6	0.9	1.3
Beluga	1.7	1.4	3.8
Prey	0.6	0.2	1.3

Table 3
Previous measurements of Cs and K on the Porcupine caribou herd (Macdonald et al., 2007) and this work in Bq kg^{-1} (ww = wet weight, dw = dry weight). Also from this work, radioactivity in mushrooms (*Russola* sp. and *Lactarius* sp.), and in lichen (*Cladina rangiferina* and *Flavocetraria cucullata*).

Bq kg^{-1}	^{137}Cs			^{40}K			
	n	Geometric mean	Geo standard deviation	Range	Geometric mean	Geo standard deviation	Range
Caribou 1990 by [Macdonald et al., 2007] (ww)	8	59	2.18	16–139	101	1.13	84–114
Caribou 2009 pre Fukushima (this work) (ww)	20	18.8	1.6	9.4–46	80.1	1.4	26–116
Caribou 2011 post Fukushima (this work) (ww)	14	9.1	1.8	3.6–20.2	76.4	1.2	42–98
Mushrooms Pre Fukushima (ww)	6	10.2	2.3	2.2–35	80.97	1.22	63–107
Mushrooms Pre Fukushima (dw)	6	90.8	2.22	27–337	723	1.22	517–976
Mushrooms Post Fukushima (ww)	6	9.1	1.9	0–19	71.99	1.09	40–99
Mushrooms Post Fukushima (dw)	6	72.5	2.04	0–173	583	1.43	362–929
Lichen Post Fukushima (ww) ^a	11	5.17	2.18	0–13.8	13.7	1.25	–13–30
Lichen Post Fukushima (dw) ^a	11	10.7	1.81	0–25	26.7	1.79	–110–61

^a For animals K is regulated in the body of the animal. In plants this may not be the case, so it is possible for plants to have no ^{40}K . Since ^{40}K is in the background which is subtracted and gamma spectrometry is a statistical process, it is possible for the background to be slightly lower (within statistics) resulting in a negative value for the amount of ^{40}K .

3.2. Beluga and beluga prey sample results

In the measurements of the beluga muscle tissue and the whole fish (beluga prey) individual samples, neither ^{134}Cs nor ^{137}Cs was above detection limits (see Table 2). However, if one summed up all the spectra from the post Fukushima beluga samples (as in Fig. 2), one finds that there was a trace amount of ^{137}Cs , namely $0.63 \pm 0.23 \text{ Bq kg}^{-1}$ (ww). If one summed up all the spectra from the pre Fukushima beluga samples, then ^{137}Cs was not found. For the beluga prey a similar analysis of summing up the spectra was

done and in the pre Fukushima prey samples $0.14 \pm 0.07 \text{ Bq kg}^{-1}$ ww was found. No ^{137}Cs was observed in the post Fukushima prey samples.

For the beluga samples, the geometric means of the measurements of ^{40}K were 87.2 and 89.2 Bq kg^{-1} ww for pre and post Fukushima samples respectively. These values are, again, close to our control value of 100 Bq kg^{-1} (ww). The geometric standard deviations were also 1.2 and 1.1 Bq kg^{-1} ww respectively and the range of values in the samples was 65.4–112 Bq kg^{-1} ww, and 68.8–108 Bq kg^{-1} ww respectively. There were no other unusual

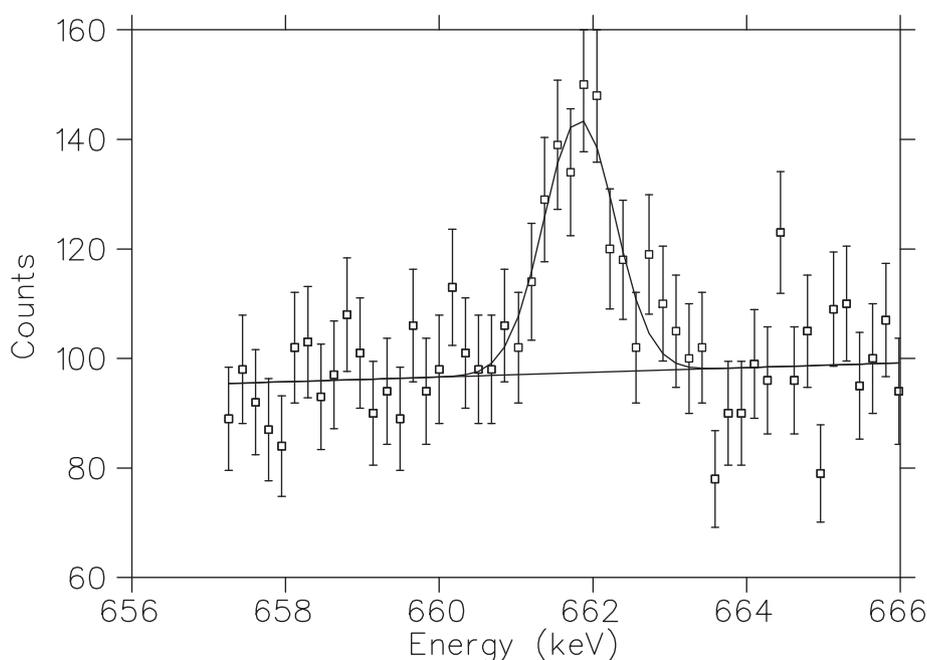


Fig. 2. The signal from Cs-137 from the summation of the spectra from beluga post Fukushima.

(anthropogenic) radionuclides found in the individual spectra. The naturally occurring radioactive material was not quantified, except for the ^{40}K .

For the beluga prey samples, the geometric means of the measurements of ^{40}K are 96.3 (pre) and 80.7 (post) Bq kg^{-1} ww. These values are, again, close to our control value of 100 Bq kg^{-1} (ww). The geometric standard deviations were also 1.1 and 1.1 Bq kg^{-1} ww respectively. The range of values in the samples was 74–118 Bq kg^{-1} ww and 66–99 Bq kg^{-1} ww. There were no other unusual (anthropogenic) radionuclides found in the individual spectra. The naturally occurring radioactive material was not quantified, except for the ^{40}K .

4. Discussion

4.1. The caribou, mushroom, and lichen

If one assumes that the ratio of ^{134}Cs – ^{137}Cs directly after the Fukushima accident is 1:1 (see for example Friese et al., 2013), one would expect that both these radionuclides would be present in the sample if the radiocesium observed was from that particular reactor accident. If one decay corrects that 1:1 ratio, taking into account the time between the Fukushima accident and the last sample to be measured, the ratio becomes 0.45: 1. Since only ^{137}Cs was observed, it is most likely attributable to the atmospheric nuclear weapons tests of the 1960s. In Table 2, the minimum detectable concentrations (MDCs) have been calculated for the various sample media for both ^{134}Cs , ^{137}Cs . Assuming this ratio of 0.45:1 and that the ^{134}Cs activity concentration is at the MDC level, one finds the amount of ^{137}Cs attributed to Fukushima under these assumptions (see Table 2). In other words, the amount of ^{137}Cs attributed to Fukushima must be less than these values, given that ^{134}Cs has not been observed. The measurements preceding this work on the same caribou herd in the early 1990s by Macdonald et al. (2007) are shown in Table 3. Comparing these results to our results here, we find that there is a decrease, as one would expect if there was no major effect from the Fukushima nuclear accident. The next step would be to see if our results are consistent with the effective half-life (which is a combination of the physical half-life and the environmental removal process) of ^{137}Cs .

By using the measured values for the activity concentrations for just the Porcupine caribou herd from Macdonald et al. (2007) and the values in this study, and by grouping the geometric means of the activity concentrations by year (see Fig. 3), we can estimate the effective half-life of ^{137}Cs in the environment for the Porcupine

herd. The estimation is done by fitting these data (Fig. 3) to an exponential function:

$$C(t) = Ae^{-\ln(2)t/T_{\text{eff}}} \quad (1)$$

where $C(t)$ is the concentration in caribou muscle as a function of time, A is the concentration at time zero, t is time in years, and T_{eff} is the effective half-life in years. Using this, the effective half-life in the environment for this herd is found to be 6.9 ± 0.6 years, which is consistent with the values reported in Macdonald et al. (2007) for all Canadian herds. The two values in the figure for 2009 and 2011 are from this study. The data was fit using MINUIT (James, 1998), where each data point was weighted by its error. This result indicates that there was no effect from a new source and is further evidence that the ^{137}Cs measured in the caribou samples is from historical fallout from the 1950s and 1960s, and not from the recent accident. Skuterud et al. (2005) noticed that there seemed to be two groups of results of this effective half-life in reindeer in the literature. One group had an effective half-life of about 3–5 years (Åhman and Åhman (1994), Amundsen (1995), Åhman et al. (2001)) and another in northern Fennoscandia which had an effective half-life of about 6–9 years (Åhman et al. (2001) Westerlund et al. (1987)). Our result agrees with the second group, which makes sense because the Porcupine herd lives at the same latitude as northern Fennoscandia. The explanation is due to the shorter growing season in the north which tends to increase half-lives. This explanation is consistent with the work of Taylor et al. (1985), which looked at effective half-lives in lichens as a function of latitude in Canada.

In the work of Hofmann et al. (1993), as in other studies on lichen sampled in Canada, a dramatic increase in the amount of ^{137}Cs was observed after the Chernobyl accident. The amount of ^{137}Cs measured in this study is consistent with, or lower than, pre Chernobyl levels measured in lichens (Hofmann et al., 1993; Rissanen and Rahola, 1990; Svoboda and Taylor, 1979; Taylor et al., 1988; Hutchison-Benson et al., 1985; Rissanen and Rahola, 1989) at various locations. This indicates that there was very little impact from the Fukushima accident on the lichen samples measured in this study. It also gives us a good baseline for comparison. In previous studies, some lichen samples showed evidence of a recent atmospheric test (Svoboda and Taylor, 1979), through the detection of the following short lived radionuclides: ^{144}Ce , ^{141}Ce , ^{103}Ru , ^{95}Zr and ^{95}Nb . Of those isotopes, only ^{144}Ce has a long enough half-life (285 days) to have been present in the samples in our study. In our work ^{144}Ce was not found in the samples. The absence of this short lived radioisotope in our samples is further evidence that there was very little impact from the Fukushima accident on the Porcupine caribou, the lichens and the mushrooms in the corresponding area. ^{144}Ce was included in the source term estimated by WHO (2012).

The Chernobyl accident showed the potential importance of mushrooms as a source of ingested radiocesium (AMAP, 2004). The comparison between our results and the AMAP shows that the levels of ^{137}Cs in mushrooms from the Yukon are much lower than in other parts of the world. The lack of ^{134}Cs in our samples, and the low level of ^{137}Cs , shows that there was no effect from Fukushima on the mushrooms sampled.

The Health Canada recommended action levels for the ingestion of contaminated food (Health Canada, 2000) for ^{137}Cs is 1000 Bq kg^{-1} . Concentrations found in caribou, beluga, beluga prey, and mushrooms from this study are well below this level.

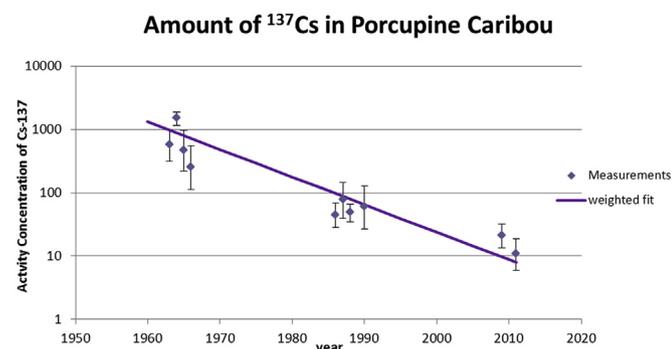


Fig. 3. Determining the effective half-life of ^{137}Cs in the home range of the Porcupine caribou herd using our results (two data points near 2010), and data from Macdonald et al. (2007). The solid line is the fit to the data, and the error bars are 1 σ error bars. The error bars for this figure were based on the geometric standard deviations, with appropriate error analysis applied. The activity concentration is in Bq kg^{-1} and the samples were taken from caribou muscle.

¹ The guideline does not specify wet weight, fresh weight, or dry weight. It should be matched to the intake rate units.

Since both Macdonald et al. (2007) and our study found little and no effect of Chernobyl and Fukushima respectively, we conclude that the ^{137}Cs currently in Porcupine caribou is from weapons fallout from the 1960s. Although we do not have measurements of Cs deposition in the home range of this herd, using estimates of global fallout as a function of latitude (Table 6 of Annex E of UNSCEAR, 1982), we can estimate deposition rates for Cs^{137} as 1.42 kBq m^{-2} pre Fukushima and 1.37 kBq m^{-2} post Fukushima in this area (between 60 and 70° north). Note that these values are 1000 times larger than the estimate deposition from Fukushima. T_{ag} values were subsequently calculated for lichens, mushrooms and caribou and were found to be consistently lower than those reported in the literature (AMAP, 2004) (Table 4). If one uses the results for T_{ag} for lichens previous to this work, one finds that for a deposition of 10 Bq m^{-2} (Health Canada, 2015), one gets values between 1 times and 5 times the MDC for ^{134}Cs (Table 2).

Since lichen and caribou samples were taken in the same year, it is possible to estimate concentration ratios, which are simply the ratio of the activity concentrations of the caribou (ww) to the lichen (dry weight - dw) or caribou (ww) to the mushroom (dw) (Leppänen, 2014; Howard et al., 2009). These concentration ratios have been calculated and are presented in Table 5. Again, these concentration factors, like the aggregate ones, change with time. Our values are smaller than the concentration factor for elemental Cs in caribou determined by Sheppard et al. (2010) which has a value of 3.1.

Finally, by using the results presented in Fig. 6 in the paper by Allaye-Chan et al. (1990), one can estimate the intake rate of lichens for August and September for the Porcupine herd and average that number to find 2.2 kg d^{-1} for lichens. Using the activity concentrations measured in this study for ^{137}Cs in both lichens and caribou, one can calculate the transfer coefficient which is

estimated at 0.55 d kg^{-1} for lichens to caribou. This estimated value assumes the caribou only receives Cs from lichens. This is a similar value to the fractional transfer (fm) calculated by Jones et al. (1989) in reindeer. That group found $\text{fm} = 0.65 \text{ d kg}^{-1}$ (January through April), $= 0.36 \text{ d kg}^{-1}$ (July), $= 0.29 \text{ d kg}^{-1}$ (August), $= 0.24 \text{ d kg}^{-1}$ (September). Our value is also slightly smaller than the elemental Cs transfer coefficient value determined by Sheppard et al. (2010), which is 0.89 d kg^{-1} .

4.2. Beluga and beluga prey

For the prey, neither ^{137}Cs nor ^{134}Cs was detected in the individual spectra. When the spectra (for all the prey species) were added for the pre Fukushima case, a small amount of ^{137}Cs was found $0.14 \pm 0.07 \text{ Bq kg}^{-1}$ ww. This value is 4.5 times lower than the value for the beluga so clearly the beluga prey are not a health risk to people.

In Cooper et al. (2000) they measured ^{137}Cs at $1.17 \pm 0.76 \text{ Bq kg}^{-1}$ dw (or 0.38 Bq kg^{-1} ww) in all tissues (blubber, epidermis, muscle, kidney, and liver) of beluga from North Slope Borough and from Resolute. These values are of the same order of magnitude as found in this research. Interestingly, in that same paper they mention that in the Beaufort Sea, the activity concentration of ^{137}Cs in water was between 1 and 4 Bq m^{-3} (0.001 – 0.004 Bq kg^{-1}). This is similar to the levels measured in 2008 by Smith et al. (2011) of 1 – 5 Bq m^{-3} (0.001 – 0.005 Bq kg^{-1}), depending on the depth in the Canadian Basin.

Like the results of Cooper et al. (2000), if one divides the measurements from this research (beluga and beluga prey) by the activity concentrations in sea water from the measurements in 2008 by Smith et al. (2011), one obtains concentration factors of 3 orders of magnitude. Cooper mentions that these 3 orders of magnitude

Table 4

Aggregate transfer factors for caribou, lichens, and mushrooms. Values in brackets are estimated average deposition (kBq m^{-2}).

Study	Lichens ($\text{m}^2 \text{ kg}^{-1}$)	Mushrooms ($\text{m}^2 \text{ kg}^{-1}$)	Caribou ($\text{m}^2 \text{ kg}^{-1}$)
This work pre Fukushima (2009)	–	0.0071 ± 0.0016^b (1.43)	0.0131 ± 0.0011 (1.43)
This work post Fukushima (2011)	0.0038 ± 0.0016 (1.37)	0.0066 ± 0.0015 (1.37)	0.0066 ± 0.0013 (1.37)
Reindeer Lom [Skuterud et al., 2014] autumn 2011		(summer)	(summer)
Reindeer Lom [Skuterud et al., 2014] winter 2011			0.039 (15)
Reindeer Lom [Skuterud et al., 2014] autumn 1987			0.023 (12)
Reindeer Lom [Skuterud et al., 2014] winter 1986			0.18 (27)
Reindeer from multiple districts [Åhman et al., 2001]			0.62 (22)
Reindeer multiple herding districts [Åhman 2007]			0.15 to 1.87
Reindeer multiple herding districts [Åhman and Åhman 1994]			0.23 to 1.07 (depending on season)
Lichens Vaga [Skuterud et al., 2005]	$(4.0 \pm 14) \times 10^{-3}$		0.12 to 0.76 (depending on season)
IAEA 2013 ^c	0.3–0.5		
Ramzaev et al., 1991 ^d	1.0		
IAEA (2010)	1.4		
IAEA 2013 ^e	0.2–2.0		
Russula spp. Kola peninsula AMAP 2004 (1992–2000)		0.029 ± 0.002	
Mezen & Nenets AO regions AMAP 2004 (1992–2000).		0.0160 ± 0.0032	

^b Error assuming 0.01 kBq m^{-2} on UNSCEAR value (none was quoted) and using geometric standard deviation.

^c Based on data from Hanson 1982.

^d Also in IAEA 2013.

^e Based on data from Hofmann et al., 1993.

Table 5
The concentration ratios (CR) (fw dw⁻¹) for caribou/lichen and caribou/mushroom.

	Geometric mean of CR	Geometric standard deviation of CR
Pre Fukushima caribou/mushrooms	0.207	3.8
Post Fukushima caribou/lichens	0.85	3.6
Post Fukushima caribou/mushrooms	0.125	3.9

are consistent with partitioning coefficients (Kd) found in laboratory experiments. This, however, is not the same as the results of Kasamatsu and Ishikawa (1997), which had concentration factors of one or two orders of magnitude in marine fishes. They, however, explain that concentration factors of three orders of magnitude can occur for Cs in sea water and biotic concentrations in the highest trophic levels, and beluga are high-level predators.

5. Conclusion

The activity concentrations in important matrices (lichens) and sources of food (caribou, beluga, mushrooms and beluga forage fish) in the north have been measured in samples collected before and after the Fukushima accident in 2011. There was no indication of ¹³⁴Cs in any of the samples. Measurements of ¹³⁷Cs in caribou were consistent with fallout from weapons testing from the 1950s and 1960s. Measurements of ¹³⁷Cs in lichens and mushrooms, in combination with the caribou measurements, yielded calculations of aggregate transfer factors and transfer coefficients. Measurements in individual samples of beluga and beluga prey did not show any ¹³⁷Cs; however, when the spectra of the post Fukushima samples were summed together, a trace amount of ¹³⁷Cs was detected which was consistent with predictions of ¹³⁷Cs from weapons fallout in the Arctic waters. The results of this study showed that there was no increase in radioactivity in these caribou or beluga as a result of the atmospheric plume from the Fukushima nuclear accident. A followup study is needed to determine if there is any affect from the oceanic Fukushima plume of radioactivity.

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