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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.

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 (^{137}Cs), which has a half-life of 30 years and is a surrogate for potassium in biological systems, easily accumulating in plants and animals. ^{137}Cs 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), Pálsson 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}Cs was 415 TBq (Health and Welfare Canada, 1987), dividing this by the area of Canada 9.98 $\times$ 10^6 km^2, 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.