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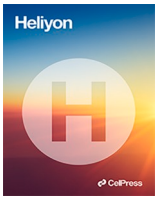

Review article

Radionuclide contamination in Canada: A scoping review

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ABSTRACT

Radionuclides were first discovered in the late 1800s, and artificial (anthropogenic) radionuclides in the 1930s. Since then, this group of substances has been increasingly incorporated into various peaceful and non-peaceful applications across Canada and the world, bringing with it both advanced technological and medical benefits, and public concern about the dangers from radiation exposure. As such, a breadth of research on, and monitoring of, radionuclides in the Canadian environment has been generated, the results of which span decades. However, a recent comprehensive review of these is not readily available. This study aims to fill this gap by synthesizing available literature from the last 30 years on the Canadian state and provenance of radionuclide contamination to better understand the context of overall sources and status of contamination. The findings indicate that while regional and temporal variations exist, on average, routine radionuclide exposure in Canada is generally attributed mainly to natural sources and fallout from historical nuclear weapons testing and nuclear accidents (including the Chernobyl and Fukushima power plant accidents) and to a smaller degree to emissions from nuclear facilities, including active and historical uranium mines and mills, nuclear research facilities, and nuclear power plants. Levels of anthropogenic radionuclides in the Canadian environment have declined since the initial cessation of nuclear weapons testing in the 1960s and are generally below guidelines protective of human health. On the national scale, present-day nuclear sector facilities do not appear to be a significant source of routine anthropogenic, nor technically-enhanced naturally occurring radionuclide exposure, though local scenarios may vary. These findings contribute context for evaluating the sustainable management of nuclear technologies, radioactive materials and waste in Canada and globally, in line with UN Sustainable Development Goal 12 and target 12.4: responsible management of chemicals and waste.

1. Introduction

Radionuclides were first discovered in the late 1800s, and artificial (anthropogenic) radionuclides in the 1930s [1]. Since then, radionuclides have been increasingly incorporated into various peaceful and non-peaceful technologies across Canada and the world, including applications in energy production, medicine, lighting, historical/tracer analyses, and weapons [2–6]. Entwined with these technological applications and advancements has been public fear of the dangers from radiation exposure due to both nuclear weapons and from peaceful radionuclide use/possession for activities such as routine power generation [7]. Together, these advancements and fears have driven the formation and direction of key regulatory bodies and frameworks to govern the use of radionuclides. For example, the International Atomic Energy Agency (IAEA) and the United Nations Comprehensive Test Ban Treaty (CTBT) on the global

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scale, and in Canada, the Canadian Nuclear Safety Commission (CNSC), its overarching *General Nuclear Safety and Control Act (NSCA)* and ancillary regulatory mechanisms, all of which aim to manage the environmental, human health and security risks from nuclear technology [6–8].

Concern over radioactivity exposure has also driven a large body of research and monitoring in Canada, on a range of natural and anthropogenic radionuclides in many environmental compartments, including air, water, sediment, soil, terrestrial and aquatic biota, and a variety of foods. The results of these efforts span decades and are largely in the public domain, via open access research papers and/or public datasets; however, while several older, focused reviews exist (e.g., [9]), a recent comprehensive review of these works is not readily available in the literature. This study aims to fill this gap by synthesizing radionuclide research and monitoring in Canada to inform the Canadian state and provenance of radionuclide contamination and to better understand the context of overall sources of concern. As a result, this context will inform the measurement of progress towards the United Nations Sustainable Development Goal (UN SDG) 12 and target 12.4 regarding the sound management of chemicals and waste such that impacts to human and environmental health are minimized [10]. This study is also a prologue for further investigation of radionuclide emissions specifically from Canadian nuclear facilities.

2. Background: radionuclide characteristics and sources

This section discusses background information on radionuclide sources to situate the later discussion on the status of radionuclide contamination in Canada (section 4). Further information on radionuclide characteristics is provided in the **Supplemental Information**.

2.1. Sources

Over 3000 radionuclides are known [11]. Many occur naturally; some stemming from when the earth was formed and include primordial radionuclides Potassium-40 (a non-chain radionuclide) and Thorium-232, Uranium-238 and Uranium-235 (and their alpha and beta decay chain products including Radon-222 and Radium-226, also called secondary radionuclides), among others [3,11–13]. Some have also been known to form underground in Canada by in situ nuclear reactions [12,14,15]. Human and non-human biotic exposure can occur due to their naturally occurring proximity in the environment (e.g., rock, air, etc.) or natural events (e.g., volcanic eruptions, wildfires, etc.). Others occur naturally when cosmic radiation passes through Earth's atmosphere (such as Carbon-14, tritium (Hydrogen-3), Sodium-22, Beryllium-10 and Beryllium-7, etc.) [1,16]. These naturally occurring radionuclides (also called, NORM - naturally occurring radioactive material) exist at very low concentrations in the natural environment, though concentrations can be technically enhanced by human activities (i.e., they are displaced/concentrated), from both within and outside the nuclear sector. Called TE-NORM (technically enhanced NORM), examples include uranium mining, which yields uranium and thorium decay chain progenies including Radium-226 and Radon-222 that are found in enhanced concentration in mine tailings. The nuclear energy sector can also contribute to enhanced levels of natural tritium, Carbon-14, as well as uranium decay chain series progenies (among other man-made radionuclides) in the environment surrounding reactors, fuel (re)processing plants and waste disposal [3,13,17–19]. Beyond the intentional use/handling of radionuclides, certain other non-nuclear sectors can also contribute to technically enhanced sources of NORMs because of industrial processing of natural materials containing them. For example, phosphate mining, fertilizer production and use in agriculture can yield enhanced levels of uranium and thorium decay chain progenies in the fertilizer product and waste stream [20]. Coal mining and combustion, oil and gas production, metal mining and smelting, mineral sands, building industry and recycling can also enhance levels of naturally occurring radionuclides [13,19,21–23], though there is some evidence that Canadian coal deposits contain relatively low amounts of radionuclides [24].

By contrast, many of the other known radionuclides are produced artificially (anthropogenic radionuclides) via bombardment of elemental nuclei (with protons or neutrons) resulting in reaction and/or fission products that are not naturally occurring [1]. Anthropogenic radionuclides are produced using nuclear reactors, particle accelerators, or radionuclide generators, and importantly, are also achieved during nuclear weapons explosions/testing [1,17]. Resulting radioisotopes include Iodine-129, Iodine-131, Molybdenum-99, Technetium-99, Xenon-133, Cesium-137, Cesium-134, Strontium-90, Plutonium-239, among others (including some overlap with primordial radionuclides), and can be used for various applications depending on their properties (e.g. half-life, radioactivity, fissionability, and chemistry), such as nuclear power, medical and/or imaging technology purposes, agricultural purposes (fertilizers, pesticides), environmental tracers, etc. [1]. Further discussion of radionuclide sources and exposures specific to Canada follows in section 4.

3. Scoping review approach

A scoping review approach [25] was taken to generally characterize the volume and focus of the body of literature related to radionuclide levels in the Canadian environment. The concepts described by PRISMA (Preferred Reporting Items for Systematic reviews and Meta-Analyses) [26], were used and comprised an online search by one reviewer using the terms "Radionuclides" and "Canada" in major online reference databases including Scopus and Web of Science. Only peer-reviewed journal articles published between 1991-present (2022), for which the full paper was accessible, pertaining to measurements of radionuclides in environmental or human samples, were retained. Further searching on the Government of Canada's Open Data Portal [27] was conducted (same search terms) to identify open access datasets produced by government programs and scientists. Several known monitoring works were also included. Table 1 describes the details of the search and selection process.

Although not always applied explicitly in the studies reviewed here, to contextualize the large body of data reviewed here, CNSC screening level guidelines [28], or Health Canada drinking water quality guidelines [29], or CODEX Alimentarius guidelines [30] have been gathered and applied to the data, where possible (i.e. radionuclide and matrix were appropriate to do so). These are described in more detail in the SI.

4. State of radionuclide contamination in Canada

The Canadian radionuclide data from research and monitoring works reviewed here include measurements in air, precipitation, water, soil, sand, sediment, various biota (birds, fish, terrestrial and marine mammals, vegetation, etc.), and food across Canada as far back as the 1950s up to present day. Fig. 1 summarizes the works reviewed in this study, excluding those for which data was not amenable to incorporation, such as, the data was not geo-tagged, etc.). In cases where exact sampling site locations were not available, locations were approximated based on available information (e.g., maps, descriptions) and the interested reader is referred to the original works for the most accurate location details. An interactive format is available (see caption link), which allows the reader to tailor the view using filters on parameters such as: year (of measurement), substance, matrix, reference, province, and whether applicable guidelines have been exceeded. The reader is encouraged to visit this dynamic version of Fig. 1 to visualize topics discussed herein. Only radionuclides are discussed here, whereas many of the studies and monitoring programs mentioned in this review also measure a range of non-radionuclide substances and parameters. The review is organized geographically based on the study areas, as follows: Canada-wide, West coast, Prairies, Ontario and the Great Lakes, Arctic, Quebec, and the Maritimes.

4.1. Canada-wide research and monitoring

Though most research studies examined here focused their investigations on targeted regions, several studies took a pan-Canadian view to radionuclide measurements in birds, air, water, milk, and vegetation. For example, Stocki & Braune [31] recently conducted analysis of historical Cesium-134 and 137 measurements in game birds (e.g. seabirds, geese, swans and various ducks, spanning a variety of habitats and dietary behaviours) during a 1989–1995 Canada-wide survey to better understand the source (i.e. whether from global fallout from historical (pre-1963) nuclear weapons testing, or from the Chernobyl nuclear accident in 1986), baseline concentrations in various trophic levels, and to assess consumption safety. Overall, they found no Cesium-134 (which is not produced by weapons tests, and with a half-life ~2.1 years, would indicate recent emission from an anthropogenic nuclear fuel source, such as Chernobyl at that time), suggesting that the source of Cesium-137 was older and likely from historical weapons testing. They found that Cesium-137 values were higher in the Hudson Bay lowlands, the Canadian Shield and the St. Lawrence lowlands relative to other Canadian areas but that most of the pooled measurements were <3 Bq/kg fresh weight (fw) (lower than levels in birds near known contamination sources). All measurements were below the CODEX food safety guideline for Cesium-137 (1000 Bq/kg fw) and thus they assessed that there was no evidence of human health risk.

Kelly et al. [32] investigated harbour radionuclide levels from several Canadian forces ports (east coast and west coast) visited by nuclear powered/nuclear capable vessels to understand whether these contributed to above background radioactivity exposures. In this follow-up to a similar 1999 investigation, Kelly et al. found that of the radionuclides they investigated (Barium-140, Cerium-144, Cesium-134/136/137/138, Iodine-131/132/133/134/135, Lanthanum-140 and Molybdenum-99) none have been observed above detection limits for any of the seawater samples analyzed over 2003–2012, and only Cesium-137 was detected occasionally in ocean sediment above the detection limit (of 1 Bq/kg dry weight (dw)). Because no Cesium-134 was observed, these findings suggest that the nuclear vessels are not a major radionuclide emission sources. Some detection of Iodine-131 on the west coast was noted in the weeks following the Fukushima- Daiichi disaster, attributed to this *trans*-pacific accidental source, whereas consistent high levels were detected in Halifax harbour kelp (generally between 10 and 100 Bq/kg dw, but reaching as high as 15,000 Bq/kg dw) were found and attributed to medical therapy sources, and not nuclear vessels. No screening level for the protection of human health was identified for Iodine-131 in aquatic plants. Earlier work by Létourneau et al. [33] adds knowledge to the source attribution of radionuclides in Canada, finding that trends (1950s–1993) of Carbon-14, Cesium-137, Gross beta activity, Strontium-90 and Tritium in air, water and milk peaked in the mid-1960s across Canada and were attributed to fallout from atmospheric nuclear weapons testing. Levels declined rapidly since the partial international cessation of aboveground weapons testing, but for two spikes in 1977 and 1981 (attributed to detonation events), and a spike representative of the nuclear accident at Chernobyl in 1986. Fig. 1 contains an excerpt of data from this study, focusing on two locations from the Ottawa Valley only, but which were deemed by the authors as representative of the overall findings for Canada.

Table 1
PRISMA flow chart for the scoping review. Counts highlighted in yellow make up the resulting body of works included in this review.

Step	Count
Initial search: Online search using terms: "Radionuclides" and "Canada" in major online databases including Web of science, Scopus, Pubmed central.	2987
First Refinement: Retained only results that were in english, peer-reviewed journal articles, for which the full paper was accessible online, published between 1991 and present, excluding those in the discipline category called 'medicine'.	477
Second refinement: Retained only studies that pertained to quantification of radionuclides in Canadian environmental or human samples. Removed duplicates.	75
Third refinement: Addition of datasets from Government of Canada's Open Data Portal, discovered using search term "Radionuclides"	14
Fourth refinement: Addition of known works via the CNSC website, Ontario Geological Survey, etc.	4

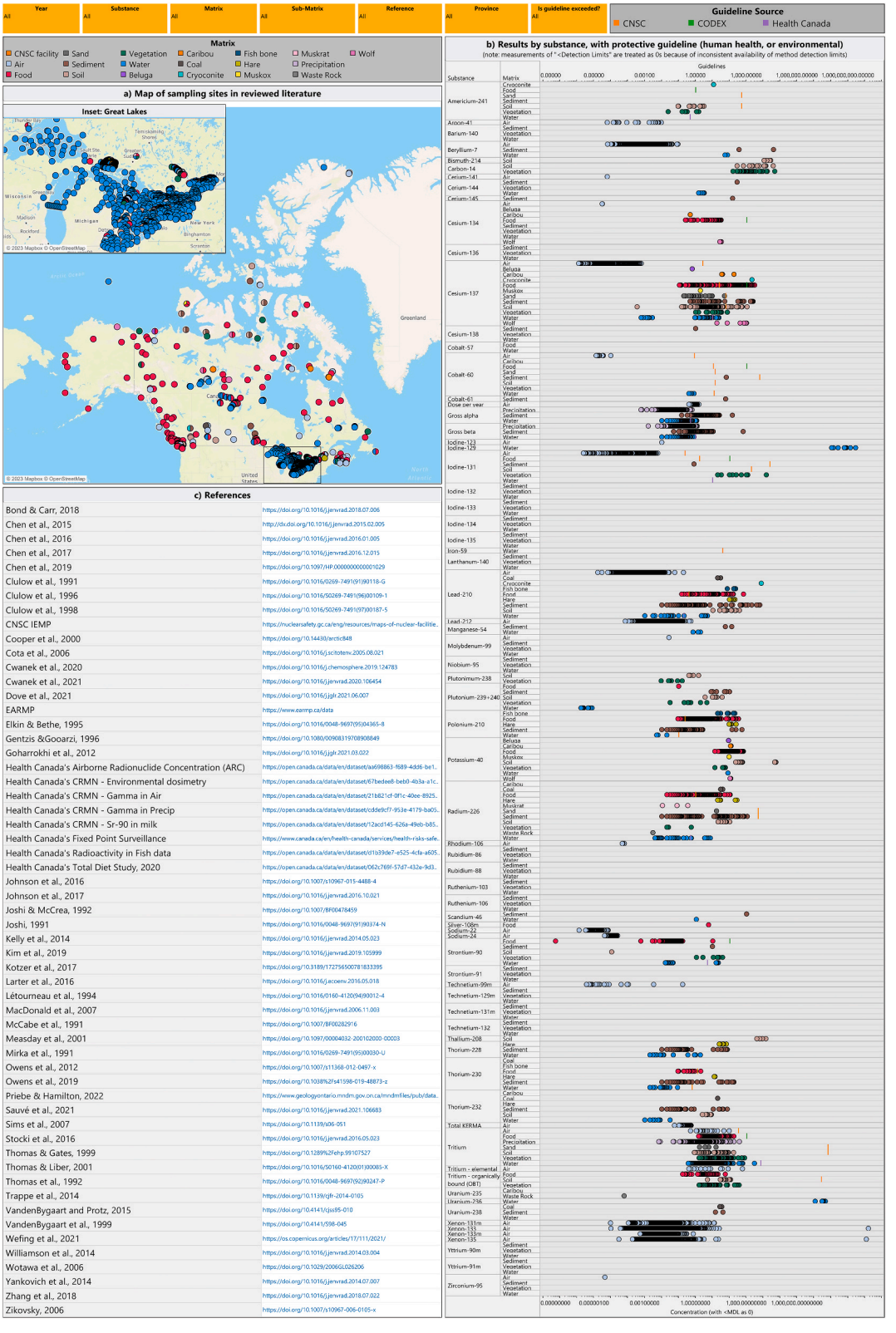

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Fig. 1. Summary of Canadian radionuclide research and monitoring data reviewed in this study. Pane a: locations of sampling sites, colour coded by the environmental matrix sampled. Pane b: concentrations per site, per year with associated guidelines protective of human health (colour-coded by guideline source). Pane c: study reference information (lead author, and DOI). **Notes:** 1) Method detection limits were not provided in all studies, therefore non-detects are represented as blanks here. 2) For studies where location data were not specified or data was not provided in a tabular format, it was re-created to the best of the author's ability using site names/description, and data surrogates based on text description of results. This was done to be as inclusive as possible in the graphical representation of the literature review, but for precise data, please see the original research papers. 3) An interactive version of this figure is available at: https://public.tableau.com/app/profile/alicia.berthiaume/viz/RadionuclideemissionsinCanada_Figure1SummaryofRadionuclideResearchandMonitoringinCanada/Figure1#1(For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Beyond the research studies, several pan-Canadian monitoring programs contribute knowledge about the state of radionuclide contamination in Canada. Of these, the broadest is Health Canada's Radionuclide Monitoring Program which comprises several efforts under its umbrella that combine routine and non-routine monitoring in various sample types at over 100 locations in Canada to inform on radiation events, sources, safety and compliance to international agreements [34]. The Canadian Radiological Monitoring Network (CRMN), the Fixed Point Surveillance Network (FPS), the Comprehensive Nuclear-Test-Ban Treaty (CTBT) Monitoring, and the Canadian Total Diet Study, are routine programs under Health Canada's umbrella, and inform on various radiations exposure routes and on radiation safety overall.

For example, the Fixed Point Surveillance (FPS) network, established in 2000, is a network of ~80 monitoring stations across Canada in both urban centers, proximal to nuclear power generation facilities, and at strategic locations along Canada's northern border [35]. It provides continuous monitoring of public gamma radiation exposure through measurements of noble gases (Argon-41, Xenon-133 and Xenon-135; responsive indicators of anthropogenic air release from nuclear power plants and some other nuclear events) and total external gamma radiation (or KERMA – Kinetic Energy Released in Matter) on a 24/7 basis at temporal resolution ranging from 24 h down to 15 min [36,37]. This data serves as an early detection system of increased anthropogenic radioactivity exposure in Canada, be it from domestic or international sources and informs emergency planning and response to these radiation events for the Federal Nuclear Emergency Plan [38], as well as providing an assessment of total and average Canadian exposures to all terrestrial gamma radiation (be it anthropogenic or natural). This data is made publicly available by the Government of Canada [35], and in addition to informing Canadian needs in this regard, the data is also fed into international efforts on radiological data exchange led by the European Commission (EURDEP), in near-real-time to inform international emergency planning and response, and is publicly available as an interactive map of radiation intensity by location [39]. Fig. 1 depicts the approximate FPS monitoring sites, along with the noble gas total air KERMA data for 2021 only, however further years of data are available online [35]. FPS data show that in 2021 Argon-41 and Xenon-133 levels were very low across Canada, ranging from 6.0×10^{-6} to 9.4×10^{-3} mSv, as was total air KERMA for most sites (average for 2021 = 0.3 mSv/year). This is about 3× lower than the 2021 average exposure measured by Health Canada's environmental dosimetry monitoring program which measures ambient background radiation doses from sites across Canada, and shows that background levels across 2016–2021 range from 0.0011 to 0.0043 mSv/day, equating to an average of ~0.9 mSv/year, attributed mainly to natural radiation of terrestrial and cosmic origin, with local variability due to properties of the location and soil, as well as seasonal fluctuations. The difference between the FPS and the environmental dosimetry datasets discussed here is explained in part by the inclusion of all cosmic radiation in the latter, versus exclusion of cosmic radiation in the former, and in part because of the nuances not accounted for here in converting total air KERMA from nGray to mSv doses. However, Liu et al. [37] have recently developed methods to include all cosmic radiation in FPS measurements and have leveraged these new methods to use the FPS to evaluate the annual effective dose of natural radiation (cosmic and terrestrial) received by the Canadian population at 36 densely populated areas across the country [40]. They found that over 2016–2020, the effective radiation dose to humans from all sources (terrestrial and cosmic) and accounting for both indoor and outdoor exposure times (89% and 11%, respectively, based on 2016 Canadian Census statistics) at these urban sites was a total of 443 μSv/year, well below the global average of 2.4 mSv/year for these natural sources [18,41,42]. In addition to the nuances between the FPS and environmental dosimetry discussed above, the accounting by this study of time spent indoors (89%) versus outdoors (11%) by the average Canadian may help explain the difference in exposure estimations in this work and data from the environmental dosimetry data discussed above.

The FPS network was also able to characterize the magnitude and extent of radioactive contamination emanating from the Fukushima-Daiichi nuclear accident and showed that airborne radioactivity was detectable in Canada for about 2 weeks in 2011 following the accident before declining below detection, and total air KERMA indicated no perceptible change from background radiation levels [43–46] also leveraged the FPS to determine that a spike in Xenon-133 at the Yellowknife sampling site in 2006 was caused by a North Korean nuclear weapons test (a non-signatory to the Comprehensive Test Ban Treaty). The FPS can also inform regional differences in average gamma radiation and in 2021, average total KERMA was highest in the Northwest Territories, followed by Yukon, whereas lowest averages were found in the Nunavut.

The Canadian Radiological Monitoring Network (CRMN) was established in 1959 to better understand the nature and extent of radionuclide contamination in Canada, including source attribution, this network comprises 28 monitoring stations (some that overlap with FPS) that represent background locations and locations proximal to nuclear sector activities like power generation [34]. Air samples are measured on a weekly basis for gamma-emitting radionuclides such as Beryllium-7, Cesium-137, Iodine-131, and Lead-210 [47] and milk was previously (1984–1993) measured for Strontium-90 annually across Canada, and is now measured only for Ottawa [48]. These data inform on both the presence and intensity of naturally occurring radionuclides, as well as human exposure potential from anthropogenic activities like nuclear power generation, fuel processing, medical applications, and nuclear weapons testing. Fig. 1 shows the pan-Canadian CRMN (approximate) sites and concentrations per year, calculated from the publicly available

data and indicate that, at the national scale, radionuclide levels in air are low and relatively stable across 2009–2022, dominated by naturally-occurring Beryllium-7 and Lead-210. Iodine-131 and Cesium-137 are orders of magnitude lower than the applicable CNSC guideline protective of health (0.228 Bq/m^3 and 2.56 Bq/m^3 respectively). Milk data on Strontium-90 indicate a steady decline nationally across the 1984–1993 dataset, continuing decline for the Ottawa-only dataset up to 2020, and well below the applicable CODEX guideline for Strontium-90 in food. Recent precipitation data (2020–21) provide measurements of gross alpha, gross beta, tritium and gamma radiation (Beryllium-7), and will be published in 2023 (replacing previously published pilot study data), however preliminary data suggests these are in the range of non-detect, <0.2 Bq/L, <7.1 Bq/L, and <6 Bq/L respectively (pers. communication, J.-F. Mercier, Radiation Protection Bureau, HC; [49]. Overall, comparison of the CRMN data in air and milk with the applicable guidelines for the protection of human health indicate that none of the measurements from the nearly 40 years of data has posed a significant risk to human health.

Similarly, Health Canada's Total Diet study [50] which measures radionuclides (Americium-241, Cesium-134, Cesium-137, Cobalt-57, Cobalt-60, Iodine-131, Lead-210, Potassium-40, and Radium-226) and other contaminants in a range of retail outlet foods and drinking water from nine cities across Canada on a systematic but non-routine basis, indicates that all foods and water measured between 2000 and 2020 are well below the available guidelines protective of health and/or not detectable above detection limits. Tritium in water vapour from sampling sites near nuclear power generation facilities is also measured by Health Canada [51] but are not shown in Fig. 1 due to lack of sampling location information in public datasets. Samples proximal to the now shutdown Gentilly nuclear generation facility in Québec [52] ranged between values below detection up to 1400 Bq/m^3, with a median of 26 Bq/m^3 for the period of 2000–2022, below the CNSC guidelines for Tritium in air of 340 Bq/m^3. For samples proximal to the Point Lepreau nuclear power facility in New Brunswick, activity ranged from values below detection limits up to 50 Bq/m^3, with a median of 5 Bq/m^3 for the same time period, the Toronto site ranged from 2 to 181 Bq/m^3 (median 35), and the Ottawa site ranged from values below detection limits to 185 Bq/m^3 (median 5); all well below the CNSC guideline for tritium in air. A more focused effort in Ottawa only looks at Tritium levels in municipal drinking water from several water treatment plants [53] and shows that tritium activity levels range from non-detect to 12 Bq/L (with many non-detects) with a median of 5 Bq/L across 2009–2021 (not shown in Fig. 1 due to a lack of location data). These measurements are all well below the Health Canada guideline for tritium in water of 7000 Bq/L, and the proposed limit of 20 Bq/L by the Ontario Drinking Water Advisory Council [54]. Fig. 2 summarizes the Health Canada routine pan-Canadian monitoring programs as well as the and targeted fish monitoring programs (discussed below in section 3.2), with associated guidelines for the protection of human health (when available), and shows that median annual concentrations have not

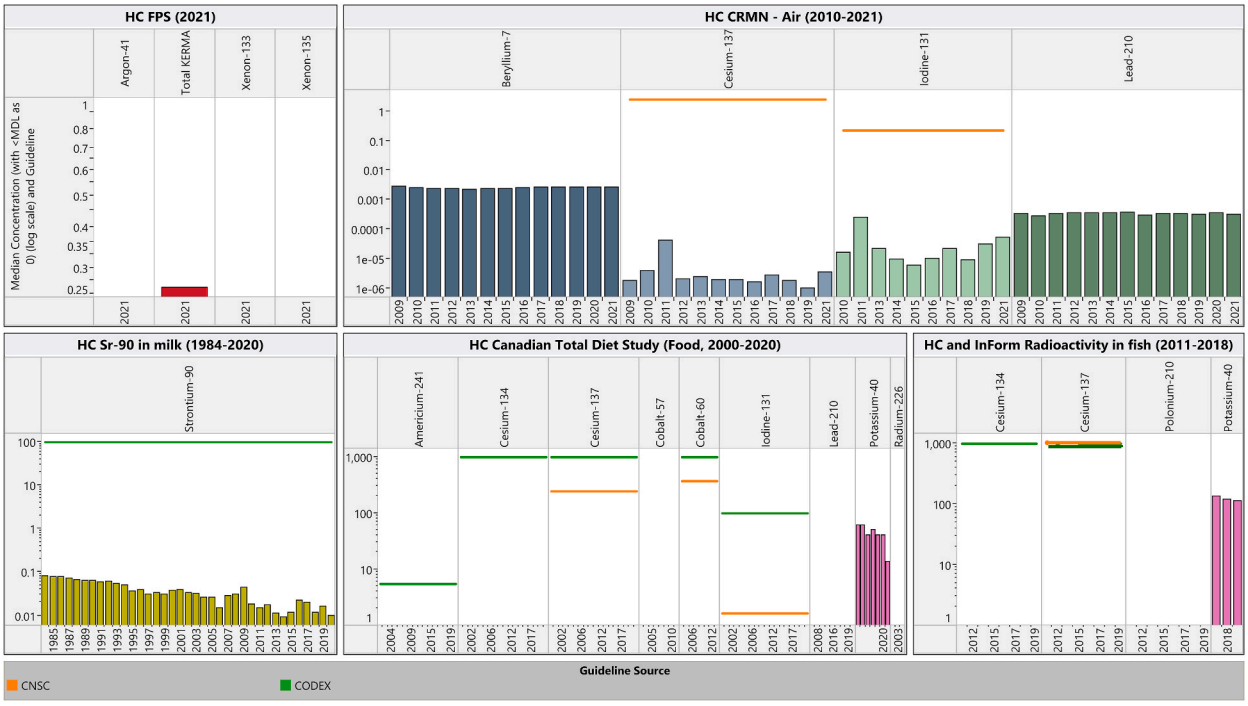

Fig. 2. Summary of Health Canada's Canadian Radiological Monitoring Network (CRMN) data available from the Government of Canada's Open Data Portal. All graphs show median concentrations per year of data available (bar charts, colour-coded by substance) with associated guidelines protective of human health (colour-coded by guideline source). Top left –right: The 2021 Fixed Point Surveillance data [35], and the CRMN air data (2010–2021) [47]. Bottom left to right: the CRNM Strontium-90 in milk data (1984–1993 for pan-Canadian sites, and 1994–2020 for Ottawa only) [48], the Total Diet study (2000–2020) [50] and the targeted fish monitoring program on the west coast of BC [62]. Note: Method detection limits were not provided in all studies, therefore non-detects are represented as blanks in this figure. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

exceeded their respective guidelines.

The Canadian portion of the international monitoring efforts for the Comprehensive Test Ban Treaty (CTBT), and is achieved through four certified radionuclide monitoring stations in Resolute, NU, Yellowknife, NT, Sidney, BC, and St. John's, NL, and a certified radionuclide laboratory in Ottawa, ON. Raw data for this program are not publicly accessible, but key analyses and maps are available from the CTBTO website [8,34].

Outside Health Canada's efforts, the CNSC conducts monitoring work across Canada, though the focus is not ambient exposure. Rather, it serves as an independent verification and public engagement tool to ensure impacts from CNSC licensees and their radionuclide releases are within safe levels for human health and the environment in the immediate and extended areas surrounding these facilities [55,56]. Called the Independent Environmental Monitoring Program (IEMP), this program provides publicly available data on a range of radionuclides (Americium-241, Cesium-137, Cobalt-60, Iodine-131, Iron-59, Lead-210, Polonium-210, Radium-226, Thorium-230, Tritium, as well as gross alpha and gross beta radiation) and non-radionuclide contaminants (not discussed here) relevant to Class I nuclear facilities. These include uranium mines and mills, nuclear power generation sites, nuclear fuel processing facilities, and nuclear labs. While a monitoring program by name, it is worthy to note that the IEMP is not designed as a fixed frequency program. The sampling schedule is irregular, and while the dataset spans 2011–2021, many facility-specific sampling campaigns are shorter, meaning that trends analysis using this dataset alone is not always possible. For example, IEMP data for the Whiteshell laboratories in MB is only available for 2017, Nordion Ltd. (Ottawa) data is only available for 2016–2018, and several of the northern SK uranium mines and mills have limited data (2014 and 2021 for Key Lake mill and McArthur River mine; 2016 for the McClean Lake operation). The dynamic version of Fig. 1 shows data for the IEMP and comprises measurements in air, sand, sediment, soil, vegetation and water, as well as local food. The location of the CNSC licensee is also shown, as are the CNSC screening guidelines. Fig. 1 indicates that almost all of the IEMP measurements of radionuclides are below the corresponding CNSC screening level, with the exception of Polonium-210 levels in food (fish) at 3 sites: the McClean Lake operating uranium mill (2016), the decommissioned uranium mine at Cluff Lake (2017), and the operating uranium mine at Cigar Lake (2020), all of which exceed the 1 Bq/kg fw CNSC guideline for Polonium-210 in fish. However, the CNSC does not consider these measurements to be indicative of licensee-caused elevated risk because these data fall within the range of background/naturally-occurring levels typical of non-impacted sites in these regions, which also produce guideline exceedances [28,56,57]. Hamlat et al. [58] looked at IEMP data from nuclear power plants in 2013 and 2015, and confirmed that measured concentrations of key radionuclides from this sector (tritium, Cesium-137, Iodine-131, as well as gross alpha radiation) were well below CNSC screening guidelines and the overall regulatory dose limit, and therefore releases from this sector were not expected to result in public exposure of radiation that would lead to health impacts. IEMP monitoring complements required on-site monitoring by licensees, which is reported to the CNSC and often available on company webpages but not discussed here.

Overall, pan-Canadian research and monitoring shows that a range of radionuclides are detectable across the country, however, few of these exceed the available guidelines protective of human health. Moreover, sources of these radionuclides are generally attributed to (in descending order) natural sources, fallout from historical nuclear weapons testing, and emissions from nuclear accidents. Based on the pan-Canadian data, current-day nuclear facilities do not appear to contribute significantly on a national scale to radionuclide exposure in Canada.

4.2. West coast

Much of the radionuclide research and monitoring on the west coast has focused on tracking and responding to the Fukushima-Daiichi nuclear accident in Japan in 2011. As noted above, the initial Canadian arrival of airborne radioactivity from the Fukushima accident was detected in the weeks following the release. And though 40 TBq of Cesium-137 were estimated to be deposited in Canada as a result of the accident [59], the initial airborne plume was characterized as a negligible increase in exposure to the average Canadian, as measured by total KERMA from Health Canada's FPS network [36,43]. Herod et al. [60] were also able to measure the pulse of Fukushima radionuclide arrival by measuring pre- and post-Fukushima Iodine-129 levels in west coast precipitation, finding an initial pulse 6 days after the accident, and subsequent increases of this isotope in groundwater samples about a year later. However, according to Smith et al. [61], the arrival and spread of the Fukushima oceanic plume on the Canadian continental shelf occurred by June 2013, ~2.3 years after the accident. In the interest of quantifying these impacts, Health Canada established a Radioactivity in Fish monitoring program to investigate levels of released radionuclides and background indicators of naturally-occurring radionuclides (Cesium-134/137, Polonium-210, Potassium-40) in various fish from the west coast of Canada (2011–2014), [62]. Beyond 2014, the work continued as a multi-stakeholder effort called InFORM (including citizen scientists from 13 west coast First Nations communities, Health Canada, and others) to continue to inform seafood safety [63]. As seen in the dynamic version of Fig. 1, sampling sites extend across the length of the west coast (and one reference site on the east coast) and comprise various mussels, scallops, oysters and their shells as well as various fish including bass, pollack, salmon, hake and chum. No samples measured were above the detection limits for Cesium-134, (indicator of Fukushima contamination) and Cesium-137 data are below the available guidelines for protection of human health from consumption of these foods, indicating that fish and shellfish from the west coast of Canada continue to pose no radiological health concerns [43,64]. These findings are re-iterated in a recent paper by Cooke et al. [65] who further delve into the 2011–2018 dataset, and is consistent with findings from the World Health Organization on fish caught outside Japan post-Fukushima [66]. Chen et al. [67] furthered conclusions drawn from this work by looking at Polonium-210 levels in salmon along the west coast, leveraging the InFORM network, and found that while radioactive cesium from the Fukushima disaster was not detectable in most salmon samples (from between 2013 and 2016), naturally occurring Polonium-210 was detected in almost all samples in levels ranging from below the detection limit of 0.2 Bq/kg fw up to 4.7 Bq/kg fw. However, the corresponding estimated annual dose from ingesting

this salmon was estimate to be a fraction of natural background dose, and therefore deemed by the authors to pose no health concern. Trappe et al. [68] extended research on Fukushima impacts by looking at Cesium-134 and 137 levels in edible mushrooms, leaf litter and soil along the west coast of Canada and the US in 2014. While levels of both isotopes were detectable in most samples, indicating at least some Fukushima-related contamination (though existing contamination was also discussed as a contributing factor), mushroom values were well below the CODEX guidelines for these isotopes in food (1000 Bq/kg fw) and the US FDA intervention level of 1200 Bq/kg (of total Cesium 134 + 137).

Prior to the Fukushima accident, Measday et al. [69] analyzed data from the Vancouver-based monitoring station for the international CTBT monitoring network. While they did not detect any non-compliances to the CTBT over the study period of 1996–1998, they did detect low-level presence of Cesium-137 attributed to background levels remaining from the 1986 Chernobyl nuclear accident, Iodine-123 attributed to releases from the local commercial isotope production facility (Nordion), Technetium-99 which is of concern insofar as it can emanate from nuclear weapons testing but in this case was attributed to emissions from medical applications at a local hospital (the Acute Care Unit of the University of British Columbia hospital); Sodium-24, Bromine-77 and several others attributed to experiments at the proximal TRIUMF nuclear research lab. Moreover, Measday et al. [69] also acknowledged low-levels of naturally-occurring radionuclides (Lead-212, Beryllium-7, Potassium-40, etc.) in air at this Vancouver station.

Also in the west, but further from the coast, Owens et al. [70] confirmed the presence of Cesium-137 in soils in two watersheds in Kamloops. Though their aim was to conduct sediment source attribution studies using fallout radionculides as a fingerprinting tool, their work was relevant here in showing that Cesium-137 levels in the BC interior soils were above CNSC screening guidelines for soil (58.6 Bq/kg dw) following wildfires. More recently, Owens et al. [71] confirmed an elevated presence of nuclear fallout radionuclides (Cesium-137, Lead-210 and Americium-241) in glacier “dirt” (cryoconite) but found that sediment in meltwaters downstream of the glacier did not contain correspondingly high levels, suggesting limited impacts to downstream aquatic environments. The level of Cesium-137 found in cryoconite was well below the 37,300 Bq/kg dw guideline for sediment protective of human health.

In summary, on the west coast of Canada, current radioactivity exposure is low and in general comes from naturally-occurring radionuclide presence, with lingering contributions from the Chernobyl and Fukushima-Daiichi accidents that may be enhanced and/or re-circulated due to wildfires, and small historical contributions from local nuclear licensees.

4.3. Prairies

Canada is one of the world’s largest producers of uranium, all of which currently comes from a cluster of mines in the Athabasca basin in northern Saskatchewan, though several new ones are being proposed in Quebec, Labrador and Nunavut, and several historic ones exist in Ontario. The mined uranium is used either domestically or exported to meet nuclear-fuel requirements of nuclear-powered electric utilities [72]. The licensee-focused monitoring in northern Saskatchewan conducted by CNSC as part of the IEMP indicates that none of the five facilities currently in operation (Cigar Lake mine, Key Lake mill, McArthur River mine, McClean Lake mill and Rabbit Lake mine and mill) or the local decommissioned site (Cluff Lake) are resulting in uranium decay chain radionuclide levels (lead-210, radium-226 and thorium-230) in the environment above corresponding screening levels protective of human health (see Fig. 1). However, fish samples from mine and mill-impacted and non-impacted reference sites near both the Cluff Lake and Cigar Lake facilities have shown Polonium-210 values above the screening level guideline (1.22 Bq/kg fw) in 2017 and 2020, respectively. The CNSC notes that in both cases, these values are within the natural background of the regions and are not expected to cause radiological health impacts beyond this natural background level [28,56]. It is worthy to note that uranium is monitored in its chemical form because as noted in section 2.3, it is more chemically toxic than radiologically toxic. The interested reader is referred to the Canadian federal government’s risk assessment of releases of radionuclides from nuclear facilities (impact on non-human biota) [73] and the associated follow-up risk management reports [74,75].

Complimentary to the CNSC IEMP in this region is the community-based Eastern Athabasca Regional Monitoring program [76]. The intention of this program is to engage the local community in monitoring changes from baseline conditions and cumulative impacts from past, present and forseeable future uranium mine and mill activities, with a focus on the safety of regional traditionally harvested food for consumption. As shown in Fig. 1, seven sites from across the Eastern Athabasca region (downstream of uranium projects) have been sampled for water and country food (i.e. fish, moose, caribou, hare, grouse, cranberries and blueberries). Samples have been analyzed for Lead-210, Polonium-210, Radium-226 and Thorium-230 as well as a range of non-radiological parameters. Data from 2011 to 2021 show that none of water samples exceeded or even approached Health Canada drinking water guidelines. However, several food samples do show some instances of measurements above the CNSC radiological screening guidelines. Moose samples from Uranium city, Wollaston lake, Stony rapids and Camsell portage exceeded the Lead-210 and Polonium-210 guidelines at several points over 2011–2021, whereas lake whitefish exceeded the Polonium-210 guideline at Wollaston in 2019 only. Despite this, the EARMP use multiple regional reference datasets to determine the range of natural background levels for these radionuclides, and departures from it, and in this context, overall, EARMP Polonium-210 measurements were within these natural background levels [77,78]. This is consistent with the IEMP findings and the 2018 EARMP human health risk findings that risks from radionculides are negiligible and that these traditional country foods are safe to consume [78]. Similar community-driven monitoring efforts are underway in the central mineral belt of northern Labrador, aiming to characterize baseline environmental conditions prior to the initiation of uranium mining activities proposed for the region [79].

Earlier work in northern Saskatchewan by Thomas & Gates [80] also showed significant levels of Polonium-210 in caribou (mean of 14 Bq/kg fw in muscle) near Wollaston, SK in 1995, though these were lower than concentrations in other regions of Canada where there are natural uranium outcrops but no uranium mining activities. This is consistent with general knowledge that caribou, especially the barren-ground caribou monitored here, are particularly prone to radionuclide exposure due to the significance of lichen in

their diet, which is an efficient accumulator of airborne radionuclides such as Polonium-210 (via Radon-222) among others [81–83]. Levels of other uranium chain decay radionuculides were also detected in caribou, and attributed mainly to natural uranium outcrops with some contribution from mining activities. Results also showed presence of Cesium-137 (attributed to nuclear weapons testing fallout) in levels below the CODEX guideline but above the CNSC guideline for this radionuclide in beef (245.7 Bq/kg fw, used here as a proxy for a lacking caribou muscle guideline) and detectable levels of naturally-occurring Potassium-40 in caribou. While radiation doses from caribou consumption (0.85–1.7 mSv/year depending on consumption patterns, mainly attributed to Polonium-210) in this area increased the lifetime risk of fatal cancer slightly, these were found to be 10× lower than elsewhere in Canada where natural concentrations of Polonium-210 and/or caribou consumption patterns were higher. Thomas & Liber [84] looked at the impact of Rabbit Lake mine on sediment levels of uranium decay chain products (Thorium-230, Radium-226, and Polonium-210) (in 1997 samples) and found elevated levels closest to the outflow of uranium mine effluent, with lower levels downstream and lowest levels at the control site, while also finding that total absorbed doses in benthic organisms at the impacted sites was 20 times higher that the control site. The study pointed to the need for further work to delineate effect thresholds for uranium decay chain products in benthic organisms to better understand ecosystem impacts from uranium mining activities.

Other work in the Athabasca region, in this case near the Alberta oil sands, by Shotyk et al. [85] looked at peat bog levels of Lead-210, leveraging Cesium-137 and Americium-241 to help date peat cores. This work strongly suggested that oil sands operations have not contributed to lead nor radionuclide levels in the area, which have declined in peat bogs since the late 1960s, effectively down to 0 in 2016 – a pattern consistent with global fallout trends. Beyond Athabasca, recent work by Lemieux et al. [86] in Alberta's Red Deer river basin investigated levels and provenance of NORM (total uranium and thorium) to better understand the distinction between anthropogenic and natural influences on NORM levels in this central Alberta region. They found that natural influences (e.g. intense rainstorms, subsequent erosion of badlands river banks, and groundwater-surface water interactions) impacted proportions and enhanced levels of Uranium and Thorium in the riverine matrices, while anthropogenic inputs from regional industry or land use (e.g. agriculture, oil and gas activities, chemical processing and wastewater) were by contrast, not significant influences.

Beyond NORM in Alberta, Sims et al. [87] measured and modelled the impact and environmental fate of a 1951 leaked mixture of fission products (containing Cesium-137 and Strontium-90) at the Canadian military base in Suffield Alberta. They found continued activity levels and slow migration of the radionuclides in the soil after 50 years, and predicted that at these migrations rates, they would pose no health hazard by the time they reached the water table (50 m below).

Peer-reviewed radionuclide research in Manitoba was sparse. A 2021 study by Goharrokhi et al. [88] measured Cesium-137 and Lead-210 in sediment from across Lake Winnipeg as part of a study on sedimentation sources and dynamics in this watershed. In general, the levels they detected were low in comparison to the CNSC sediment guideline for Cesium-137 (37,300 Bq/kg dw) and tended to be higher in the northern part of the lake versus the southern part. Beyond this, national monitoring programs provided measurements of both natural and anthropogenic radionuclides in various media, mainly in the southern part of the province near Winnipeg and Whiteshell Laboratories. These data indicate the presence of Beryllium-7, Cesium-137, Iodine-131, Lead-210/212/214, Potassium-40, Strontium-90 and tritium, but no guideline exceedances.

4.4. Ontario and the Great Lakes

Several research and monitoring efforts have focused specifically on radionuclide levels in Ontario, including investigations in water, sediment and food (fish). As can be seen in Fig. 1, Ontario hosts the majority of both current and historical CNSC licensed nuclear facilities. Recent focus particularly on the Great Lakes may have also arisen in part because of attention generated from a recent nomination by ENGOs to add radionuclides to the list of chemicals of concern in the Canada-United States Great Lakes Water Quality Agreement (GLWQA) [89]. The proposal cited health and environmental concerns associated with radionuclides particularly in the Great Lakes Basin that the proponent found deserving of the additional attention, and enhanced reduction measures that such an addition would afford. Concerns related to radionuclide releases from the nuclear power generation sector and ancillary sub-sectors (e.g., nuclear fuel fabrication, current spent fuel waste management), as well as historical mines, the medical sector, and potentially from future long-term radioactive waste storage. The CNSC led the Canadian evaluation of the proposal [13], and the resulting report recommended against further consideration of radionuclides as a chemical of concern in the GLWQA because knowledge at the time (research, monitoring, modelling, etc.) indicated that this group of substances were not posing an unreasonable risk to the environment, nor human health and safety in the Great Lakes region [13]. The report noted that for most anthropogenic radionuclides in the Great Lakes, levels peaked in the 1960s and these were attributed to atmospheric nuclear weapons testing, findings supported by 1990s studies by Joshi [90] and Ahier & Tracy [91]. Anthropogenic radionuclides have since decreased overall in Ontario, despite the initiation of nuclear power generation in the province in 1970 [13,90,92]. Source attribution of tritium, of concern because of its potential/actual release from CANDU (**Can**adian **D**euterium **U**ranium) nuclear power reactors, was shown to be dominated by legacy fallout from historical nuclear weapons testing, except in the vicinity of CANDU reactors, where small routine releases (few Bq) from reactors were the dominant sources [13]. Tritium levels are expected to continue to decline based on projected inputs and losses (i.e., decay of legacy sources, current CANDU emissions trends/projections, sediment sinks, and other model parameters) and remain relatively low at approximately 4 Bq/L through to 2025 [92]. Recent work by Dove et al. [93] to measure Tritium levels in all five Great Lakes over 2017–2019 and combine those with compatible historical data back to the 1990s, supports the trends discussed above. As shown in the dynamic version of Fig. 1, declines in Tritium in all Great Lakes have been observed, consistent with the decay of legacy fallout from nuclear weapons testing in the 1960s, which still dwarf present-day contributions from nuclear power facilities. Measurements made in 2017 and 2019 are orders of magnitude lower than the corresponding Health Canada drinking water guidelines (7000 Bq/L) [93] and lower than the proposed limit of 20 Bq/L by the Ontario Drinking Water Advisory Council [54].

Aside from the Great Lakes, radionuclides have been measured elsewhere in Ontario, and tend to show that natural and anthropogenic levels are below those significant to human health. For example, the Ontario Geological Survey recently conducted a comprehensive survey of tritium levels in shallow groundwater in southern and eastern Ontario over 2007–2018 [94]. The resulting interpolated "heat maps" of tritium concentrations show the highest levels to be along the shore of Lake Ontario from Toronto to Darlington (proximal to several nuclear sector facilities), peaking near Pickering, and second highest levels in Bruce County. All measurements in this study were well below the Health Canada tritium drinking water guideline as well as below the proposed and the proposed limit of 20 Bq/L by the Ontario Drinking Water Advisory Council [54]. Another example is the work by Chen et al. [95] who examined radioactivity in fish from the non-impacted Experimental Lakes Area in northern Ontario to better understand background radiation levels in country foods (see Fig. 1). While levels of naturally occurring Lead-210 and Radium-226 were not detectable, Polonium-210 was detectable in all fish, suggesting that natural background levels in this region are not insignificant and emphasizing that regional context affects interpretation of naturally occurring radionuclide measurements. Chen et al. also investigated, but did not detect, indicators of recent inputs from nuclear accidents but did detect Cesium-137, attributed to historical sources (e.g., fallout from nuclear weapons testing, the Chernobyl nuclear accident, other sources, and to a small degree, the recent Fukushima accident). Overall, however, the resulting radiation dose from consuming these fish was estimated to be a fraction of natural background radiation and therefore not of human health concern. Similar fish measurements were also conducted by Chen et al. [43] in the Ottawa Valley and Chen et al. [67] on the west coast of Canada. Comparison of these two studies showed that Cesium-137 levels (~6.1 Bq/kg fw) in northern Ontario were about twice as high as in the Ottawa Valley, and 20 times higher than concentrations in fish from the Fukushima impacted west coast in 2013 and 2014 [67], though well below the CNSC guideline of 1040 Bq/kg fw [28]. Chen et al. [95] theorize that despite similar exposures, marine fish may have experienced less Cesium-137 uptake compared to their northern Ontario freshwater counterparts, owing to excess potassium in seawater that can block uptake of cesium.

Work by several groups in the Ottawa Valley leveraged the unique conditions this region provides (proximity to multiple radioxenon sources from local medical isotope production in Chalk river, regional hospital medical isotope use, Lake Ontario nuclear power plants), to advance radiation detection technology and meteorological atmospheric plume models in support of real world verification of models for both local and world-wide source identification [96,97]. Such work contributes to improved accuracy of CTBT compliance verification, which relies on radioxenon detection as an indicator of nuclear tests and CTBT contraventions. For example, Stocki et al. [96] used Health Canada's FPS network in conjunction with a radioxenon analyzer in Ottawa to detect abnormal but compliant and non-health-threatening xenon pulses in air, and then verified atmospheric transport/dispersion models to attribute the source of the xenon to releases from Chalk river nuclear facilities. Johnson et al. [97] also detected large spikes in xenon releases and downwind impacts to air from this facility, confirming earlier recognition that this facility, due to its production of the medical isotope Molybdenum-99, has been a significant source of global radioxenon [44,98]. Leveraging this fact, the Chalk River site was also used as a test site to develop methodology for subsurface radioxenon detection in support of on-site-inspections for CTBT. This research found Xenon-133 in both atmospheric and underground air samples near Chalk River in 2015, and demonstrated that elevated levels of subsurface radioxenon can be detectable for days after plume passage and thus of value to on-site inspection activities [98].

Chalk River has also been a site for radionuclide research in terrestrial and aquatic systems. Earlier work published in 1992 by Joshi and McCrea [99] documented various Chalk River lab-derived radionuclides downstream in the Ottawa river, though these were very low level (in the mBq range). Strontium-90 presence was attributed to global fallout mainly, and not the Chalk River facility, whereas Cesium-137 from the lab appeared to contribute a proportion equal to the global fallout source. This investigation also looked at whether the radionuclides were subsequently removed during Ottawa drinking water treatment, and found that some were, and some (including Strontium-90 and Cesium-137) were not. Kim et al. [100] looked at the spatial footprint (in soil) of the 60 years of tritium releases to air from this facility and found levels of tritiated water and organically bound tritium in soil on the site varied geographically in line with wind direction. Maximum levels detected for both tritiated water and organically bound tritium between 2012 and 2014 were <200 Bq/L of fresh soil, well below the CNSC guidelines (HTO - 64,200,000 Bq/kg fw, organically bound tritium – 26,200,000 Bq/kg fw). And while not measured in situ, Zhou et al. [101] used lab experiments to investigate what would transpire if wildfires occurred in radionuclide contaminated forests, using vegetation from the Chalk River laboratory site. They found that while release to air of Strontium-90 from vegetation combustion is negligible (it remains in ash), the release of Cesium-137 varies with combustion temperature and vegetation type, implying that forest fires in radionuclide contaminated vegetation could recirculate previously stored Cesium-137 to air, as found in situ by Owens et al. [70] on the West coast. Similar re-mobilization results were found for radioiodine in biomass fires [102]. With the potential number and area of wildfires increasing due to climate change [103], redistribution of radionuclides as a result of fires continues to be of scientific interest, however, research in terrestrial systems as highly contaminated as those of the Chernobyl exclusions zone indicate this exposure pathway does not pose a significant public risk [104].

In southern Ontario, in the Lake Erie basin, soils examined in the 1985-89 were confirmed to contain naturally occurring radionuclides (e.g. Potassium-40 and Uranium-238 daughters), as well as Cesium-137, which were attributed to global nuclear bomb testing fallout [105]. Similarly, VandenBygaart & Protz [106] and VandenBygaart et al. [107] confirmed the same radionuclides are present in soils and vegetation (litter) in Pinery Provincial Park, and other points in southwestern Ontario, while also positively correlating the background radionuclide content with clay content in soils.

Very few of the research and monitoring works focused on Ontario have yielded radionuclide measurements above their respective Health Canada, or CNSC guideline for the protection of humans over the span of 1953–2021. In the Bancroft area, while Desbarat et al. [108] found several 2011–2012 groundwater samples at various historical uranium mines sites exceeded their respective Radium-226 and Lead-210 guidelines (0.5 Bq/L and 0.2 Bq/L respectively), water samples from beyond the fence line analyzed by the CNSC as part of the IEMP did not exceed these guidelines. Sauvé et al. [109] evaluated water and sediment cores collected in 1993 to quantify historical radionuclide trends (of Lead-210, Polonium-210, Radium-226, Thorium-228/-230/-232, Gross alpha and Gross beta

radiation) in the Serpent river watershed in northern Ontario, a site impacted by both uranium mine operations as well as natural background levels in the area (Fig. 1). When these measurements are compared to the Health Canada drinking water guidelines, exceedances arise for Gross alpha, beta and Radium-226 in water in both the exposed and unexposed riversheds. The authors acknowledge that natural background levels play an important role in determining recovery targets for environmental management of uranium exploitation activities and legacies. Also in the Elliot Lake area in the 1990s, Clulow et al. [110] looked at impacts to biota from uranium mine tailings, which at the time had been inactive for a few decades and reclaimed (chemically stabilized and revegetated) (Fig. 1). They found that snowshoe hare near the tailings sites accumulated Radium-226 in their bones at higher rates than control sites, but that Lead-210 and Polonium-210 were at similar levels at both impacted and non-impacted sites, and all levels measured were well below cancer-causing thresholds and so unlikely to be adversely affecting the hare. They also found that uranium and thorium were not accumulated in the skeletons of these animals, and that in general, snowshoe hares were not a significant dispersion pathway of radionuclides from tailings to other locations. Similar findings by Clulow et al. [111,112], and Mirka et al. [113] of higher levels of uranium chain radionuclides in beavers, muskrat, fish, cattails, water and sediment close to impacted sites (versus downstream sites and non-impacted sites) in the 1990s, Elliot Lake region, still concluded that estimated human radiation exposure via consumption of these animals was within allowable limits in Canada. Williamson et al. [114] confirmed Uranium-235 and Radium-226 presence in reclaimed waste heap piles in the area when conducting various leaching tests on these waste piles. More recently, 2017 targeted assessment of northern Ontario mushrooms as part of the CRMN found Cesium-137 ranged from 0.3 to 57 Bq/kg fw, well below the CODEX guideline of 1000 Bq/kg fw in food, while Cesium-134 was not detected (not included in Fig. 1 because a lack of sampling location information) [115].

In summary, in Ontario, exposure to radionuclides is from both natural sources and anthropogenic sources, which are mainly from legacy nuclear weapons fallout and to a smaller degree, from historical uranium mining, the Fukushima accident and emissions from nuclear research facilities and power plants.

4.5. Arctic

Radionuclide research and monitoring in the Canadian arctic have been driven largely by interest in characterizing the nature and extent of contaminants in the north, in general. Both the Canadian Northern Contaminants Program (NCP) and the international Arctic Monitoring and Assessment Program (AMAP) were established in the 1990s to support contaminant research in this remote cold-climate region (that tends to accumulate long-range contamination from both historical and current-day anthropogenic inputs from the south), giving rise to several overviews of the state and occurrence of radionuclides in the arctic (discussed below). As shown in the dynamic version of Fig. 1, the range of environmental matrices sampled in the Arctic is comprehensive, including air, water, soil, sediment, vegetation (lichen), a range of biota (beluga whales, seals, reindeer, wolf, muskox, and caribou), a range of foods, and comprises measurements of a range of radionuclides. None of the measurements in the last five decades described in these studies exceeded the available corresponding radionuclide guidelines specific to these matrices, though such guidelines are not available for all combinations presented here.

In the 1990s, Barrie et al. [116], Lockhart et al. [117] and Thomas et al. [118] described the sources and occurrence of anthropogenic radionuclides to the arctic as being mainly from atmospheric testing of nuclear weapons (prior to the signing of the 1963 international partial test ban treaty), two nuclear-containing satellite re-entry accidents (1964 - SNAP-9A satellite, 1978 - Cosmos 954), several post-treaty weapons tests by non-signatory nations (up to 1980), and the Chernobyl accident (1986). Two technically enhanced naturally occurring radionuclide sources from local uranium ore deposits in Port Radium and Rayrock were also acknowledged as much smaller and more localized than the latter atmospheric sources, and giving rise to Radium-226 and uranium in levels in nearby lakes below Health Canada drinking water guidelines [117]. However, Traditional Knowledge not previously accounted for in these areas has uncovered significant contamination and legacy effects from historical mining, and points to limitations in assessment approaches [152]. Kotzer et al. [119] recently examined trends of tritium in ice cores from Ellesmere Island (Agassiz ice cap) that corroborate Arctic sources and timeleines, and demonstrated a first phase of tritium increase in polar ice across 1946–51 (attributed to early fission bombs), and a second substantial increase during 1952–64 (attributed to thermonuclear hydrogen-fusion bomb detonation). Macdonald et al. [120] extended this same storyline by adding that nuclear fuel reprocessing facilities in Europe (England, France and Scotland) have likely also contributed to Canadian marine arctic radionuclide loads. The dumping of submarine and marine nuclear reactor assemblies containing spent fuel by the former Soviet Union in the 1970s–1990s (in contravention of the London Dumping Convention 1972 [121]) are also noted as a previously unrecognized source to international arctic marine environments, including Canada. Cooper et al. [122] also acknowledge dispersal of anthropogenic radionuclides in the Arctic Ocean and Canada Basin from Russian riverine sources via the Laptev shelf, likely related to Russian nuclear fuel reprocessing. Overall however, Macdonald et al. [120] note that the despite the anthropogenic sources, it was naturally occurring radionuclides (e.g. Lead-210, Polonium-210, Thorium-228 and Thorium 232) that made up the majority of human exposure for arctic populations in the 1990s, and that these total doses were “not large enough to warrant immediate concern”. Van Oostdam et al. [82] agree and note that the bioaccumulation of naturally occurring radionuclides (present in the arctic for millenia) in the traditional country food chain has been the most significant contributor to human radiation doses in the Arctic. However, as with the findings for the EARMP in northern SK where comparatively high natural background levels do not negate the safety of traditional country food, the resulting arctic exposures do not negate the importance and safety of traditional country food [123]. A follow-up to the former study, also led by Macdonald [124], further investigated the impact to Canadian arctic marine waters from international sources and confirmed their earlier findings. Several factors were cited to explain this low radiation risk status: overall levels of anthropogenic radionuclides in marine environments were lower than corresponding risk thresholds, reduced emissions from fuel reprocessing operations since the

1970s (and decay of earlier larger inputs), and containment in deep waters of the Atlantic Ocean inhibiting/slowing transport to Canadian territory. It was acknowledged that if containment barriers of dumped radioactive waste in northern Russian waters were to be breached, resulting contamination via oceanic transport could be expected in Canadian arctic territory within 10 years of release [124], which could change this overall assessment of risk. In 1999, Braune et al. [125] leveraged the earlier Thomas et al. [118] and MacDonald et al. [120] findings and found that radionuclide measurements in the arctic were geographically comprehensive, re-iterated the source attributions found by previous authors, and noted the continued decline of Cesium-137 in caribou since the 1960s. Moreover, recent studies by Karcher et al. [126], Wefing et al. [127] and Lopez-Lora et al. [128] recently tracked Iodine-129, Uranium-236 and others and confirmed anthropogenic inputs to Canadian arctic oceans sourced mainly from nuclear processing facilities in northern Europe and to a smaller degree, from global fallout. The former study also showed a climate-change related increase in the movement and mixing of Atlantic waters into arctic ones, suggesting that arctic radionuclide contamination from lower latitude oceanic sources may be exacerbated by climate change. Meese et al. [129] and Landa et al. [130] also acknowledged that transport of radionuclides from non-Canadian sources (e.g., nuclear fuel/weapons (re-) processing in Europe and Russia) via ice-borne sediment and ice raft debris is a means by which radionuclides are entering the Canadian arctic via the Beaufort sea. Meanwhile, Cota et al. [131] found unexpectedly high measurements of Cesium-137 in sediment from grounded ice floes in the high arctic (Resolute Bay, NU), but based on mineralogical composition matching, they could not attribute the source of these high readings to Russian sources, rather they found it difficult to ascertain the cause and source(s). Recently Miroshnikov et al. [132] have suggested that the peculiar efficiency of arctic cryoconite to accumulate radionuclides is related to the melting of a buried layer of ice that was contaminated during the age of atmospheric nuclear testing. While the maximum values measured by Cota et al. were on the order of 1800–2000 Bq/kg dw, well below the CNSC guidelines for this radionuclide in sediment (37,300 Bq/kg dw), it is not unreasonable to expect that climate change may potentially further enhance exposure levels from this secondary source of legacy contamination. Indeed, Bond & Carr [133] support this notion of climate-change re-mobilization, finding that tritium levels, while low relative to the Health Canada drinking water guideline, were higher in regions of melting permafrost (i.e. MacKenzie river valley – a region with extensive permafrost degradation due to warming climate), as compared to regions where permafrost was in tact (effectively maintaining trapped legacy tritium from historical weapons testing).

Cooper et al. [134] added to the knowledge base with radionuclide measurement in a range of marine mammals confirming the known presence of Cesium-137, but also reported the unusual finding of radiosilver (Ag-108 m), which was attributed as a bomb fallout product that bioconcentrated in local the beluga food chain. Elkin and Bethe [81] looked at various arctic caribou herds and found moderate levels of Cesium-137 (<185 Bq/kg wet weight) in the 1990s, while Macdonald et al. [83] looked at the anthropogenic radionuclides Cesium-134 and Cesium-137 in caribou and reindeer across the arctic and found that despite geographic and seasonal variations, levels declined overall in both species since the 1960s (consistent with decay and disappearance of inputs following the test ban treaty), with a detectable but minor signal for the Chernobyl accident in 1986. The authors estimate that both maximum Cesium-137 doses in caribou in the 1960s (e.g. 2000–3000 Bq/kg wet weight) and 9× lower doses in the 1990s correspond to radiation exposures below levels at which acute harm to the animals is expected. Though not done so in the study, it can be inferred that the levels in caribou in 1990s were within today's Cesium-137 CODEX guideline for food safety (1000 Bq/kg), though if the CNSC screening level for beef (245 Bq/kg fw) is used as a surrogate for a lacking caribou guideline, then there were many exceedances. The reindeer data yielded similar findings of maximum concentration in the 1960s (903 Bq/kg fw) with a downwards trend into the 1990s, and all data within the CODEX guideline for food safety, but only below the CNSC screening level for beef in the 1990s and not before (see dynamic version of Fig. 1). Human body burdens of Cesium-137 across the late 1960s to 1990 were analyzed by Tracy et al. [135] and indeed show that radiocesium doses had declined in residents of five Canadian arctic communities by about two orders of magnitude, averaging 0.04 mSv/year by 1990, which was attributed to both declining exposures from caribou, as well as a change in the pattern of traditional country food consumption.

Radionuclide measurements by Cwanek et al. [136,137] in arctic freshwater systems (sediment and soil sampled in 1999) and terrestrial vegetation (lichen sampled in 2012) support that the above findings hold true for inland environments as well, noting that in general, anthropogenic radioactive pollution (i.e. Americium-241, Cesium-137, Plutonium-238/239 + 240 and Strontium-90) of arctic freshwater systems is present but low (and varies based on geography and meteorology) (see dynamic version of Fig. 1). Prevailing anthropogenic sources noted here are in line with previous works: global fallout from historical weapons testing, and to a smaller degree the Chernobyl nuclear accident pre-2011, with evidence of Fukushima contaminants in western arctic lichen post-2011. And while they also corroborate the findings that anthropogenic radionuclides comprise a smaller fraction of overall radioactivity exposure relative to NORM, and lower than more temperate environments, they also found evidence of resuspension of radionuclides in previously immobile arctic sediment, consistent with climate-change induced re-mobilization found by Bond & Carr [133] and Miroshnikov et al. [132].

Stocki et al. [138] looked at impacts to the arctic ecosystem (caribou, beluga, lichens, mushrooms, and beluga prey (fish)) from before and after the 2011 Fukushima accident and did not detect any increase in radioactivity overall, nor any Cesium-134 in the post-accident samples, indicating no recent nuclear fuel contamination. They did however find low levels of Cesium-137 in caribou, lichen, mushroom and beluga (<10 Bq/kg fw in all matrices) which they attributed to legacy fallout from historical atmospheric weapons testing and acknowledged that these low levels are well within safety limits for consumption and therefore not of radiological concern to human consumers (see dynamic version of Fig. 1). Similarly, Chen et al. [139] looked at post-Fukushima conditions in the arctic but focused on both natural and anthropogenic radionuclides in seals, and as with the latter study, found no detectable Cesium-134 and only low levels of Cesium-137 (<0.3 Bq/kg fw) which were well below the CODEX guideline for food (1000 Bq/kg fw). They concluded these data indicate no radiological health concern to consumers of seal meat. (see dynamic version of Fig. 1). By contrast, Larter et al. [140] did indeed detect Cesium-134 in a small number of mountain goat and Dall's sheep in the southern

MacKenzie mountains (NT) in 2011, 2012, and 2013 but none in mountain caribou nor moose from the same region. They note that this detection indicates at least some contamination this region from Fukushima fallout, but also noted that the overall Cesium-134, -137 levels they measured were low enough that risks to animals and human consumers were negligible. Moreover, although not done so in this study, comparison of the measured data to CNSC and CODEX guidelines for Cesium-137 in beef and both Cesium isotopes in food, respectively indicate that the measurements are well within these limits (see dynamic version of Fig. 1).

Regarding arctic air, Chen [141] discussed the seasonal variation of naturally occurring Lead-210 levels in this matrix, which were found to be higher in the winter (vs. summer) months. This difference was attributed to the Arctic Haze phenomenon, whereby inputs of NORM (and other pollutants) via long-range transport build up in winter, then scavenged efficiently in summer, creating a seasonal pattern that is the reverse of what would be expected if NORM inputs were from local origin only. The projected impacts of climate change on the long-range transport of airborne NORM and/or anthropogenic radionuclides were not part of this study and may be an area for future work. The presence and seasonal variations in natural (cosmic) radionuclides Beryllium-7 and Sodium-22 at Resolute Bay were discussed by Zhang et al. [142], where it was found that these peaked in Spring and Summer, respectively, attributed to seasonal patterns in arctic air mass mixing. Wotawa et al. [143] note that (re)contributions of legacy Cesium-137 contamination in biomass to arctic air from intra- and/or intercontinental transport of wildfire emissions may also be a factor to consider in both magnitude and frequency of radionuclide sources to arctic air. Wildfires in Alaska, Yukon and Siberia in 2003–2004 were estimated to account for Cesium-137 re-suspension in air in amounts in the TBq range in those years, yielding measurement in Yellowknife, NT of 1–15 $\mu Bq/m^3$ during the burn periods. Though these levels were well below the CNSC guidelines for Cesium-137 in air (2.56 Bq/m^3), given the expected increases in wildfires due to climate-change, this source may become more important in the future.

In summary, in the arctic, exposure to radionuclides is present but generally low, and occurring due to both natural sources and anthropogenic sources, the latter of which are mainly from legacy nuclear weapons fallout and to a smaller degree, from European oceanic and Asian atmospheric long range transport, and the Chernobyl and Fukushima accident. Climate change is influencing the mobility of legacy radionuclides trapped in biomass, sea ice and permafrost, with the effect of enhancing exposures from these previously immobile contaminants.

4.6. Quebec and the maritimes

Research on radionuclide levels in Quebec and the Maritimes are sparse though national monitoring programs provide coverage of these regions. In Quebec, a drinking water study spanning 20 years (1984–2004) indicated presence of Radium-226 at an arithmetic mean of 0.0044 Bq/L, well below the Health Canada guideline of 0.5 Bq/L [144]. Beyond that, Health Canada and IEMP monitoring programs for air, precipitation, soil, vegetation, and food (including grocery store items as well as reindeer and caribou) collectively indicate presence of both natural and anthropogenic radionuclides over 1984–2021 in these regions, but no guideline exceedances. Previously discussed high Iodine-131 findings in Halifax Harbour kelp by Kelly et al. [32] suggest medical isotope use in the region may be a source of regional significance. See Fig. 1.

5. Discussion and conclusion

In summary, over 75 journal articles and 16 published monitoring program datasets were reviewed here comprising over 150K data points in ~200 environmental matrices and food-types, and over 65 radionuclides from across Canada. This review was undertaken to establish the state and provenance of radionuclide contamination in Canada and to establish context for evaluating progress towards the sustainable management of radioactive materials and waste both domestically and globally, in line with UN SDG 12 and target 12.4: responsible management of chemicals and waste [10]. This work also serves as a prologue for further investigation of radionuclide releases reported by Canadian nuclear facilities. This analysis has shown that while regional and temporal variations are significant, natural (primordial, secondary, and cosmic) and anthropogenic radionculides are somewhat ubiquitous in the Canadian environment. In Canada, anthropogenic radionuclide contamination is generally attributed to fallout from historical nuclear weapons testing and nuclear accidents (crashes of nuclear technology-containing satellites, Chernobyl and Fukushima power plant accidents), and to a smaller degree from emissions from nuclear facilities, including active and historical uranium mines and mills, nuclear research facilities, nuclear processing facilities, and nuclear power plants. Generally, sources beyond these, though possible, were not reflected in the literature reviewed here. Rather, this analysis emphasized the lasting effects of legacy nuclear weapons testing and nuclear accidents on the Canadian environment and demonstrates the importance of continued vigilance in minimizing such releases in the future. It is important to note that research and monitoring data not readily accessible in the public domain, such as data from the Federal Contaminated Sites Inventory [145] or other repositories (e.g. private), was not considered here, nor were indicators of contamination beyond environmental measurements (e.g., traditional knowledge, among others). This gap presents an opportunity to integrate such omitted information into the Open Data forum, to provide further insights into the history, current state and provenance of radionuclides in Canada.

Overall, levels of anthropogenic radionuclides in environmental matrices have generally been below various guidelines protective of human health (CNSC screening levels, CODEX guidelines) in the last 5 decades (1970 – present). In certain cases, levels of radionculides in particular environmental matrices (i.e. fish, soil) surpassed guidelines though these instances are related to pre-1970s data, or high natural occurrence, or to enhancement of legacy contamination from re-circulation due to wildfires. An investigation of the validity and protective intention of the myriad guidelines applied in this review was beyond the scope of this study and may merit further attention [146]. Moreover, the availability guidelines for all relevant radionuclide/matrix combinations was a limitation noted, as was the general focus of more commonly used guidelines (e.g. from Health Canada, CODEX and CNSC) limited to

anthropocentric protection. Dose-coefficients for non-human biota environmentally exposed to radiation are available from the ICRP [147], but are not discussed in the research and monitoring reviewed here. Future opportunities for more ecocentric perspectives of radionuclide impacts in Canada appear ripe if the gap in availability and awareness of guidelines for the protection of non-human organisms in and of themselves, is filled. Doing so will broaden the understanding of ecosystem-wide radionculide impacts in Canada and beyond, and domestic and global progress towards sustainability in the context of the UN SDG 12.4 regarding the sound management of chemicals. Indeed, this need and concept of further integrating ecosystem science and ecological receptors in assessment of radionuclide contamination has recently been recognized at the international level [148], and is an emerging area of research that will benefit future Canadian radionuclide status reviews.

Finally, the synthesis of monitoring programs provided in section 4 provides a cohesive assimilation of many individual programs available in open access format, though many adjustments were needed to align these separate datasets (e.g., nomenclature, units, parameters, format, etc.). This incompatibility in terms of location and format of open access data from the federal government can discourage assimilation with other data towards higher interpretations of results. There is an opportunity to improve consistency and cross-talk between radionuclide data from federal programs and potentially beyond, such that their analytical compatibility is improved, in line with the principles of comparability and interoperability of Canada and G8 Open Data Charter ([149,150]).

Author contribution statement

All authors listed have significantly contributed to the development and the writing of this article.

Data availability statement

Data included in article/supplementary material/referenced in article.

Additional information

Supplementary content related to this article has been published online at [URL].

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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