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# The effect of climate change on sources of radionuclides to the marine environment

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Climate change interacts with the sources and cycling of contaminants, such as radionuclides, in the environment. In this review, we discuss the implications of climate change impacts on existing and potential future sources of radionuclides associated with human activities to the marine environment. The overall effect on operational releases of radionuclides from the nuclear and non-nuclear sectors will likely be increased interference or prevention of normal operations due to weather-related events. For certain radioactive waste dumped at sea and sunken nuclear submarines, the impact of climate change and ocean acidification on the release of radionuclides and their subsequent fate in the marine environment should be considered further. Fluxes from secondary sources of radionuclides in the marine and terrestrial environment and cryosphere will change in response to climate change impacts such as sea level rise, warming and changes in precipitation patterns. In addition, climate change impacts on the marine environment. Overall, our synthesis highlights that there is a need to understand and assess climate change impacts on sources of radionuclides to the marine environment to meet environment and management challenges under future climate scenarios.

The consequences of climate change (CC) have already led to global social, economic, and environmental impacts<sup>1</sup>. The emerging interaction between CC and the sources and cycling of contaminants, including radionuclides leads to poorly constrained impacts that affect the sensitivity of organisms to contamination, leading to impaired ecosystem function, services, and risk assessment evaluations<sup>2</sup>. Here, we examine and discuss the implications of relevant CC impacts on existing sources of radionuclides to and within the marine environment from present and past, planned and accidental human activities, and future potential sources of radionuclides (Fig. 1). Increased awareness of the scope of CC impacts on the range of existing and potential sources of radionuclides is essential for national authorities, commercial and environmental stakeholders and the wider public alike to better understand future risks of changes in the current levels of radionuclides in the marine environment. In addition, through such understanding, the need for mitigation or adaptation can be identified and addressed where and as appropriate. This review is limited to impacts on sources of radionuclides from human activities to the marine environment, but it should be noted that CC impacts, as well as ocean acidification, are likely to have a myriad of effects on all aspects of marine environmental radioecology.

In this review, existing sources of radionuclides are defined as those that are occurring at present through operational releases from industrial activities (e.g., authorized discharges from nuclear facilities), those that are already present within the marine environment (e.g., dumped radioactive waste) as well as secondary sources where historical contamination of the marine and terrestrial environment and cryosphere have occurred. Transfer to and within the marine environment will be influenced by environmental conditions, processes (chemical, physical, and biological), and the behavior of the individual radionuclides. The mobility and reactivity of individual radionuclides in the environment can vary considerably and will reflect any changes in environmental conditions. Here, it is important to remember that large inputs of radionuclides to the marine environment have occurred from past human activities (Box. 1). Potential sources of radionuclides that are considered in this study are operational and legacy wastes that exist on land and which through CC impacts may lead to additional unplanned inputs of radionuclides to the marine environment.

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Fig. 1 | Overview of observed and predicted impacts of human-caused climate change on the oceans, the land, and the cryosphere relevant for sources of radionuclides to the marine environment. Observed and predicted impacts of human-caused climate change on the oceans,

the land, and the cryosphere are taken from reports published during the IPCC's sixth assessment  $cycle^{3-5}$ . Although not a CC impact per se, impacts from ocean acidification are also included in this review.

#### Box 1 | What past human activities have led to the release of radionuclides?

Human activities have and continue to provide sources of man-made and naturally occurring radionuclides to the marine environment (e.g., fallout from nuclear weapon testing, nuclear accidents (e.g., Chornobyl and Fukushima), past and contemporary authorized discharges from nuclear facilities as well as from non-nuclear industries (mainly from the phosphate industry and offshore oil and gas production))<sup>35,41,74,220</sup>. The radio-nuclides released into the marine environment from such human activities vary considerably in terms of their physical half-life, behavior in the marine environment, potential for biological uptake, and radiological impact on humans and marine organisms.

Global fallout from nuclear weapons testing in the 1950s and 1960s has been the dominant input to the marine environment since the dawn of the nuclear age<sup>74</sup>. However, point sources such as the authorized releases from the European nuclear reprocessing facilities at Sellafield (UK) and la Hague (France) have led to higher levels of certain radionuclides in the receiving waters where these discharges have occurred and along the subsequent oceanic transport pathways as the releases have been dispersed<sup>82,221–225</sup>. In addition, accidental releases such as those from Chornobyl (1986) and Fukushima (2011) have resulted in pulsed increases in the levels of certain radionuclides in affected marine regions<sup>226–226</sup> that have then decreased over time because of dilution as well as radioactive decay. Inputs from such sources have also resulted in

Overall, our synthesis highlights that CC impacts will lead to changes in inputs of radionuclides to the marine environment. Therefore, there is a need to understand and assess CC impacts on sources of radionuclides to the marine environment to meet environmental and management challenges under future climate scenarios.

#### Climate change impacts

During the Intergovernmental Panel on Climate Change's (IPCC) sixth assessment cycle, special reports were published outlining the observed and predicted impacts of human-caused climate change on the oceans and the accumulation of radionuclides in the terrestrial environment, which have then been transferred to the marine environment over time through different environmental processes<sup>76,229</sup>. For comparison purposes, Supplementary Table 1 lists the inputs of cesium-137 (<sup>137</sup>Cs), one of the main contributors to radiological impact, to the marine environment from the main sources of man-made radionuclides (i.e., global fallout, discharges from nuclear fuel reprocessing facilities and nuclear accidents) and which have been in the order of  $10^{\circ}$  to  $10^{2}$  PBq ( $10^{15}$ Bq)<sup>74,220,230–235</sup>. Additionally, it should be noted that such sources have also given rise to inputs of a range of other man-made radionuclides to the marine environment. Smaller Inputs from other sources of man-made radionuclides have also occurred in the past that may be relevant for certain marine areas, such as the past operational discharges from the French nuclear reprocessing facility at Marcoule that discharged around 73 TBq (1012 Bq) of 137Cs (as well as other radionuclides) to the Mediterranean between 1964 to 2016<sup>236</sup> (Charmasson S., pers. comm.).

Inputs of naturally occurring radionuclides to the marine environment from human activities have also occurred in the past at levels that have been comparable in terms of activity with inputs of man-made radionuclides, most notably through the discharges of phosphogypsum waste<sup>35</sup>.

cryosphere<sup>3</sup> and on land<sup>4</sup> as well as a report on the physical science of climate change<sup>5</sup>. Although not a CC impact per se, the IPCC includes impacts from ocean acidification in the relevant aforementioned reports as the decrease in ocean pH is a direct result of the atmospheric increase in the greenhouse gas carbon dioxide. Impacts from ocean acidification are included in this review in the same regard. Due to the levels of greenhouse gases currently in the atmosphere, the trends of certain CC impacts (e.g., sea level rise<sup>6</sup> and ocean oxygen loss<sup>7</sup>) will continue. However, projections on the overall degree of all CC impacts over the remaining 21st century will depend on the scale of future greenhouse gas emissions and global warming scenarios. An

overview of the observed and projected CC impacts and their respective confidence levels, as reported by the IPCC, that have been identified as relevant for this review, are given in Supplementary Tables 2 and 3 and summarized in Fig. 1. CC impacts can individually give rise to multiple consequences, for example, sea level rise can result in increased erosion, flooding and salinization of coastal areas, or contradictory consequences such as changes in terrestrial runoff fluxes due to impacts of increases in heavy precipitation events versus reduced snow cover. In combination, certain impacts can give rise to compounding effects; for example, the impact of sea level rise and extreme wave heights on extreme sea level events and the increase in heat-related events on the increase in wildfires and dust storms<sup>5</sup>. CC impacts can be exacerbated further through socioeconomic development that results in increased demand for water resources and land use change<sup>1</sup>. It should be remembered that there is a regionality to CC impacts even where an impact has been observed globally. Intensification rates of hurricanes, for instance, have increased near the Atlantic coast of the United States (US), but not for the Gulf coast<sup>8</sup>, similarly, ocean acidification is increasing at a rate three to four times higher in the western Arctic Ocean than in any other ocean basin9.

# Climate change impacts existing sources from present human activities

#### Operational releases from nuclear facilities

All nuclear facilities can have discharges (liquid releases) and emissions (atmospheric releases) that can enter the marine environment either directly (e.g., for liquid releases from coastal facilities) or indirectly via rivers (e.g., for liquid releases from inland facilities) and atmospheric transport. Such discharges and emissions can be due to operational and/or decommissioning activities taking place over the lifetime of a nuclear facility (Box. 2).

#### Climate change impacts on operational releases from nuclear facilities

In terms of CC impacts on operational releases from nuclear facilities, reductions in discharges can be expected where such impacts interfere with or prevent normal operations. Due to the requirement for vast volumes of water for cooling as part of the power/steam generation cycle, it could be considered that NPPs are more vulnerable to CC impacts than any other type of nuclear facility. For NPPs located on rivers and lakes, CC impacts leading to low water levels and/or increased water temperatures can result in reduced output or a complete stop in electrical generation (outages) or reduced efficiency at times when energy demands can be increasing (e.g., during a heatwave)<sup>10-13</sup>. In the US, droughts between 2006 and 2012 forced numerous NPP to reduce output or shutdown reactor units<sup>12</sup>. As well as providing problems in terms of cooling, increased water temperatures can create challenges associated with the maximum temperature of returning used cooling water to avoid thermal pollution<sup>10-13</sup>. In 2003 in France, a warmer-than-average summer resulted in a total power loss of 5.3 TWh, equivalent to >200 days of reactor operations<sup>10,13</sup>. However, only an average 0.3% of annual production was lost in France due to high water temperatures and low river flows between 2000 and 202214. According to the Organization for Economic Cooperation and Development (OECD)<sup>12</sup> and using data from the IAEA's Power Reactor Information Systems (PRIS) database, warm cooling water problems accounted for 71.4% of all weatherrelated energy losses between 2004 and 2013, yet this loss was only 0.12% of the total electricity production during this period. Interestingly, the OECD stated that cold cooling water and ice formation accounted for 16% of weather-related energy losses during the same period<sup>12</sup>, an outage cause that might be expected to diminish with increased overall warming. NPPs at coastal sites maybe less vulnerable to warm cooling water and low water level problems, yet in the US, for example, coastal NPPs must adhere to similar regulations on thermal pollution as for NPPs situated on rivers and lakes<sup>12</sup>.

Although NPPs located on rivers and lakes can also be exposed to flood events (e.g., the Missouri River flooding at Fort Calhoun, Nebraska, in 2011), one of the main concerns for NPPs at coastal sites is that the risk of flooding during tropical cyclones and storm surges will be exacerbated by sea-level rise<sup>12</sup>. However, during Hurricane Sandy in 2012, of the 27 reactor units (coastal and inland) that were in the path of the storm, 24 continued to operate, albeit with some at reduced power<sup>12</sup>. Only one reactor unit was shutdown due to high water levels, while two reactors were shutdown due to off-site problems with the electrical transmission grid, which also accounted for the need for other reactors to reduce output<sup>12</sup>. The experience from other hurricanes (category 1 to 4) in the US between 2011 and 2018 is similar, with most affected NPPs able to continue operating at full power, with any requirements to reduce output or shutdown mainly due to disruptions with off-site electrical grids<sup>15</sup>. It is worth recalling that off-site electrical transmission grids are also vulnerable to other weather-related events than tropical cyclones, as occurred to devastating effect with the 1998 ice storm in Canada<sup>12</sup>.

When considering all weather-related causes of NPP outages between 2004 and 2013 that were reported to the IAEA PRIS database, the total loss of electricity production was 44.7 TWh, which only amounted to 0.2% of the total electricity that was generated by NPPs over this period worldwide<sup>12</sup>. This would suggest such outage causes have only a limited effect on normal NPP operations at present.

A further likely consequence of CC impacts on normal operations at NPPs, whether coastal or inland, is through biofouling. There have been numerous documented cases of jellyfish blocking cooling water intakes to coastal NPPs (and even to a nuclear-powered vessel) from different regions of the world<sup>16-18</sup>. Blooms of jellyfish are predicted to increase due to warmer ocean temperatures and ocean acidification<sup>19</sup>, with a recent study linking such blooms directly to the thermal pollution of returning cooling water from an NPP<sup>20</sup>. Increased river temperatures on the Ebro River in Spain have been linked to increased growth of freshwater plants that have led to a reduced capacity for the intake of cooling water and a consequent need for a reduction in electricity production at the Ascó NPP<sup>12,21</sup>. Biofouling of cooling water intakes of NPPs by invasive mollusc species (e.g., Zebra mussels (*Dreissena polymorpha*)) has been a problem for decades<sup>22-25</sup>, with suggestions that suitable habitats for some invasive species may increase compared to others under future climate scenarios<sup>26</sup>.

The IAEA's specific safety guide on meteorological and hydrological hazards for the site evaluation of nuclear installations, includes advice on assessing impacts from extreme precipitation events, storm surges, wind wave effects, and biofouling<sup>27</sup>. The guide also states that since the planned operating lifetime of an NPP is assumed to be of the order of about 100 years, the variability of and changes in regional climate should be considered, with the uncertainties in climate projections taken into account<sup>27</sup>.

The current edition of this specific safety guide references the IPCC's Fourth Assessment Report<sup>28</sup> and so refers to an anticipated rise in mean sea level of 0.18 to 0.59 m by the end of the 21st century. However, this is somewhat lower than more recent estimates made during the IPCC's Sixth Assessment cycle of a mean sea level rise of 0.43 to 0.84 m (range of 0.29 to 1.10 m) by the end of the 21st century and based on the minimum and maximum global warming scenarios under consideration<sup>3</sup>.

In many cases, NPPs have already had to develop mitigation and adaptation measures to deal with problems such as flooding<sup>29</sup> and biofouling<sup>30</sup> to minimize disruption to normal operations. Further mitigation and adaptation challenges due to increasing CC impacts may need to be addressed, especially when considering that some current and new NPPs will have lifetimes that will extend well beyond the end of the current century.

As a response to CC and a need to meet national energy decarbonization targets as well as energy security issues, some countries are currently looking to increase the share of electricity generation by nuclear power<sup>31,52</sup>. Although some currently operating NPPs are expected to be decommissioned in the years ahead, as of April 2023 and according to the World Nuclear Association<sup>33</sup> there are 60 reactors currently under construction, 99 reactors planned, and a further 334 reactors proposed worldwide with a combined total maximum output of 540 GWe, 14 times the total output in 2020. Any increase in the total number of NPPs constructed is likely to lead to a concomitant increase in the overall operational

#### Box 2 | What and how much is released from nuclear facilities?

The main types of radionuclides released through emissions are radioactive noble gases and carbon-14, whereas the releases of other types of radionuclides (e.g., tritium) are typically greater through discharges (Supplementary Table 4). Discharges and emissions are carried out under permits issued by national authorities and must comply with the International Atomic Energy Agency's (IAEA) safety standards for the radiation protection of the public and the environment (i.e., any public exposure from normal operations must be below 1 mSv per year<sup>237</sup>). However, it should be noted that there are orders of magnitude differences in the activities released and the type of radionuclides released across different nuclear industries. For example, based on data reported to the OSPAR Commission for discharges from nuclear industries to the North-East Atlantic, the two nuclear fuel reprocessing facilities at Sellafield (UK) and la Hague (France) accounted for 93%, 66%, and 83% of all discharges of total alpha, total beta (excluding tritium) and tritium in 2020, respectively, whereas NPPs contributed only 0.02%, 32%, and 17%, respectively (Supplementary Table 5). As the reprocessing of nuclear fuel at Sellafield ceased in 2022, it can be expected that the magnitude of discharges from this facility may decrease. In terms of the total activity discharged from the nuclear sector to the North-East Atlantic in 2020, 99.9% was attributable to the discharge of tritium, for which there is no currently available technique for the industrial scale removal of this radionuclide from

operational discharges produced by nuclear facilities. At the end of December 2020, there were 429 individual reactor units in operation, with a maximum total output of 38.5 GWe, at civilian NPPs around the world<sup>238</sup>. Of these 429 individual reactors, 178 are located across 70 sites directly on the coast (Box Fig. 2). The open publication of discharge data for NPPs varies between countries that use nuclear energy to produce electricity. However, averages for the different types of reactors in use can be calculated based on available data (Supplementary Table 6). Using this approach, we can estimate total annual discharges of 0.0002 TBg for total alpha, 5.2 TBg for total beta/gamma (excluding tritium), and ~9000 TBq for tritium for the 429 reactors in operation at NPPs around the world in 2020. These values are lower than the corresponding values for the discharges from the two European nuclear reprocessing plants at Sellafield and la Hague in the same year (Supplementary Table 9). These values should be considered a conservative estimate for the activity that enters the marine environment, as these include the total discharges from NPPs situated on rivers or lakes inland. The fraction of radionuclides reaching the marine environment from any discharges to rivers or lakes inland may be less than the total discharged as result of physical decay and retention within the waterways where such discharges occur.



**Box Fig. 2** Locations of civilian nuclear power plants (NPPs) with at least one reactor in operation in 2020. NPPs located directly on the coast are shown by blue circles and those inland by red squares. This figure does not include the location of the Russian floating NPP Akademik Lomonosov in the Far East of Russia, which was not included in the estimation of total annual discharges from civilian NPPs (see Supplementary Table 6), as it is not known whether operational discharges occur from these transportable reactor units. Locations of NPPs provided by the World Nuclear Association.

release of radionuclides into the marine environment from this nuclear industry.

#### Operational releases from non-nuclear industries

Operational releases of radionuclides to the marine environment in waste streams from non-nuclear industries are typically but not limited to naturally occurring radionuclides. The main sources of operational releases of such radionuclides to the marine environment from both a historical and contemporary perspective have been from the phosphate industry and offshore oil and gas production (Box. 3), although releases of naturally occurring radionuclides from other non-nuclear industries, such as the production of rare earth elements, primary steel, and titanium dioxide production have also occurred<sup>34–37</sup>. The industrial processing of phosphate rock to produce phosphoric acid, driven by the global demand for

phosphate fertilizers and animal feed, produces a range of waste products that can contain elevated levels of naturally occurring radionuclides<sup>35,38,39</sup>, and in particular <sup>226</sup>Ra in phosphogypsum. While discharges from the phosphate industry are linked to coastal areas, offshore oil and gas production results in operational releases of naturally occurring radionuclides to open seas, through the by-product discharge of produced water, which predominantly contain the radionuclides <sup>226</sup>Ra and <sup>228</sup>Ra<sup>40,41</sup>. Additionally, unintended releases of naturally occurring radionuclides to the marine environment can occur during the transport and handling of mineral ores, coal, and other raw materials intended for various industrial uses<sup>42-44</sup>. Operational releases of man-made radionuclides from non-nuclear industries also occur from the production of radiochemicals and radio-pharmaceuticals and the use of typically short-lived isotopes for therapy and diagnostic purposes in hospitals<sup>37,45,46</sup>.

# Box 3 | What is phosphogypsum waste what is produced water, and how much of these wastes are discharged?

Phosphate rock contains naturally occurring radionuclides of the uranium-238 (<sup>238</sup>U) and thorium-232 (<sup>232</sup>Th) decay series, and depending on the extraction process used, a substantial proportion of <sup>226</sup>Ra can be transferred into the phosphogypsum waste along with smaller amounts of other radionuclides from these decay series<sup>239,240</sup>. Reserves and processing of phosphate rock occur mainly in China, Morocco, the US, other African countries, the Middle East, and Russia<sup>35</sup>. Typically, there has been little potential for the further use of phosphogypsum waste, with only 10–15% of current production reutilized in construction and other industries<sup>199,241</sup>, and therefore the waste has either been stored on land or discharged<sup>35</sup>. The IAEA has reported the global discharge of phosphogypsum waste to water bodies was between 1.8 and 3.0 billion tonnes up to 2006<sup>35</sup>, with the majority of such discharges likely to have occurred directly to coastal areas. Due to tighter regulations on the phosphate industry, direct discharges in most production countries have ceased<sup>35</sup>. However, such discharges continue in some countries, notably Morocco<sup>95</sup>, with a contemporary estimated global annual discharge of phosphogypsum of 31 million tonnes<sup>35</sup>, equivalent to 31 TBq per year of <sup>226</sup>Ra, when using a global average of 1 Bq/g for <sup>226</sup>Ra in phosphogypsum<sup>35</sup>. Based on this annual discharge, derived values for total alpha and total beta annual discharges (using the approach adopted by the OSPAR Commission for deriving such values for discharges of produced water (See footnote to Supplementary Table 7)) are two orders of magnitude higher and fivefold higher than the respective discharges of total alpha and total beta from the two nuclear fuel reprocessing facilities at Sellafield and la Hague in 2020 (Supplementary Table 9). Assuming a similar annual discharge of phosphogypsum for the period from 2006 to 2020, the range of total discharges of total alpha and total beta (based on <sup>226</sup>Ra alone) up to 2020 would be 8.9 to 21 PBq and 4.5 to 10 PBq, respectively (Supplementary Table 7).

Produced water is formed by the mixing of seawater injected into the oil or gas reservoir to maintain production pressure with any in-situ formation water. The ratio of volumes of produced water to oil or gas that is extracted is not constant and increases over the lifetime of individual production wells. Although there can be controls on the amount of oil that

#### Climate change impacts on operational releases from nonnuclear industries

Regarding operational discharges of radionuclides from the phosphate industry and similar processing industries, no evident reductions would be anticipated directly from CC impacts other than potential disruptions to normal operations due to weather-related events and any such disruptions linked to off-site electricity supplies. Increases or reductions in the unintended releases of naturally occurring radionuclides to the marine environment during the transport and handling of relevant raw materials will be dependent on region-specific CC impacts, for example, through changes to mean wind speeds and the frequency and intensity of severe windstorms<sup>47</sup>.

Offshore oil and gas production is far more vulnerable to weatherrelated disruptions of normal operations, with this sector likely to face greater challenges under future climate scenarios<sup>48</sup>. Disruptions of normal operations in offshore oil and gas production have occurred in all regions affected by tropical cyclones and other severe storm systems<sup>49–52</sup>. The time taken to resume normal operations can vary from days to months, with only 90% of pre-storm levels reached for US production in the Gulf of Mexico six months after the category 5 hurricanes Katrina and Rita<sup>53</sup> in 2005 when 113 production platforms were destroyed <sup>54</sup>. Similarly, in 2008, the category 4 hurricanes Gustav and Ike destroyed 60 platforms, responsible for about 1.6% of the oil and 2.5% of the gas produced daily in the Gulf of Mexico<sup>50</sup>. can be discharged in produced water, there is no available technology to remove the fraction of naturally occurring radionuclides from what is subsequently discharged. Reinjection of produced water back into the oil and gas reservoir can be an option, with reported reinjection rates for different offshore production regions in 2020 ranging from 10% to  $100\%^{242}$ . Discharged produced water typically contains low activities of naturally occurring radionuclides, with ranges reported of 0.002 to 1200 Bq/l for <sup>226</sup>Ra and 0.3 to 180 Bq/l for <sup>228</sup>Ra<sup>40,41</sup>. However, due to the volume of produced water that is discharged, the overall input of radionuclides from the offshore oil and gas industry (as well as minor contributions from onshore cleaning of equipment) can be substantial.

Offshore oil and gas production is mainly concentrated in Saudi Arabia, Brazil, Mexico, Norway, and the US, although it should be noted that onshore oil and gas production can also result in the discharge of produced water to freshwater systems that may eventually reach the marine environment. Globally, and based on available data, although the reinjection of produced water is higher (82% in 2020) for onshore oil and gas production compared to offshore (32% in 2020), onshore production accounts for the majority (71% in 2020) of the total annual global oil and gas production<sup>242</sup>. Estimating the annual global discharge of naturally occurring radionuclides from the offshore oil and gas industry is not a straightforward task due to inherent differences in the age, geology, and type of hydrocarbon production of individual production fields that can influence the volumes of produced water and activities of naturally occurring radionuclides that are discharged<sup>243-245</sup>. Further complicating any global estimate is the paucity of data on actual discharged activities of naturally occurring radionuclides from the regions where offshore production occurs. Nevertheless, for 2020 and using two different approaches (Supplementary Table 8), estimates of the global annual discharge of naturally occurring radionuclides from offshore oil and gas production for total alpha and total beta give values that are higher than the respective discharges of total alpha and total beta from the two nuclear fuel reprocessing facilities at Sellafield and la Hague in the same year (Supplementary Table 9).

As a further response to meet national energy decarbonization targets, some countries have already pledged to phase out oil and gas production, with France agreeing to stop production by 2040 and Denmark by 2050<sup>55</sup>. However, continued reductions in Arctic sea-ice cover may allow for oil and gas extraction in the Far North and potentially any associated operational discharges of produced water. It has been estimated that the total mean undiscovered and conventional oil and gas resources of the Arctic are ~14 billion Sm<sup>3</sup> of oil and 47 billion and 7 billion Sm<sup>3</sup> oil equivalent of natural gas and natural gas liquids<sup>56</sup>, respectively.

# Climate change impacts existing sources from past human activities

#### Dumped radioactive waste

From 1946 until the international ban on the dumping of radioactive waste at sea in 1993, thirteen countries (Belgium, France, the former Soviet Union and Russia, Germany, Italy, Japan, Netherlands, New Zealand, the Republic of Korea, Sweden, Switzerland, the United Kingdom, and the US) dumped radioactive waste in the oceans<sup>57</sup> (Box. 4). Radioactive waste that was dumped at the Atlantic and Pacific sites were mostly carried out following international guidance and consisted of low-level waste dumped in containers at depths of at least 3000 m<sup>57</sup>. Of greater concern are the reactor units that have been dumped, either with or without spent nuclear fuel (SNF), by

the former Soviet Union in bays along the eastern coast of Novaya Zemlya in the Arctic at depths (20 to 300 m) far shallower than was recommended for dumping at that time<sup>58</sup>.

#### Climate change impacts on dumped radioactive waste

It is difficult to see how CC impacts might influence the ongoing slow release of radioactive waste that was dumped at the deep Atlantic and Pacific Ocean dump sites. However, ocean acidification is projected to increase in abyssal bottom waters, with global mean pH estimated to decline by  $0.018 \pm 0.001$  to  $0.030 \pm 0.002$  by the end of the 21st century based on the minimum and maximum global warming scenarios under consideration<sup>59</sup>. It is not known whether increased ocean acidification will affect corrosion rates of these dumped containers. The issue of increased corrosion rates may be more relevant for dumped radioactive waste in bays along the Eastern coast of Novaya Zemlya, as ocean acidification in surface waters is expected to increase at a higher rate than in bottom waters, especially within the Arctic<sup>5,9</sup>. It has been suggested that the containers dumped by the former Soviet Union would have already been subject to substantial corrosion 20 to 30

#### Box 4 | What was dumped and where?

years after they were dumped<sup>60</sup>. As other CC impacts that may influence corrosion rates are predicted to increase (ocean temperatures) or decrease (dissolved oxygen concentration in seawater), further work may be required to look at the effect of these changes on the issue of corrosion in greater detail. It was reported that the waste dumped in these shallow water bays by the former Soviet Union could be prone to physical damage by the seabed gouging of seasonally formed sea-ice<sup>61</sup>, but this would be a risk that might be expected to diminish under future climate scenarios.

The environmental processes that control the exchange of water within the bays along the Eastern coast of Novaya Zemlya and the open Kara Sea are likely to change (i.e., seasonal freshwater runoff, seasonal ice formation, and wind forcing) under future CC impacts. Previous modeling studies have examined the flushing times of any large-scale releases of radioactivity from some of the bays where radioactive waste was dumped and concluded that any such releases would not have any meaningful impact on the levels of radionuclides in the Kara Sea<sup>62-64</sup>. All these studies stated that there would be a strong seasonality in the flushing times, with shorter times in the summer than compared to the winter, mainly due to the presence of sea-ice cover

Upwards of 400,000 containers of solid and mainly low-level radioactive wastes at sites in the Atlantic and Pacific Oceans and the Arctic, with a total activity of 46,000 TBq<sup>57</sup> (Box Fig. 3). Of this solid low-level waste, 94% of the total activity was dumped at sites in the North-East Atlantic with most of the activity due to beta-gamma radionuclides, including tritium (Supplementary Table 10). As part of the dumping guidance and concept, the containers were designed to ensure the waste was delivered to the sea floor and then slowly corrode and release their contents over time. Studies of the bottom seawater at dump sites in the North-East Atlantic have shown small increased levels of some alpha and beta-gamma emitters, showing that measurable releases from the dumped containers have occurred<sup>56,246</sup>.

The reactor units dumped by the former Soviet Union account for ~44% of the total activity of all radioactive waste that has been dumped in the oceans (Supplementary Table 11). In addition, it has been reported

that 12,281 containers with low-level solid radioactive waste were dumped within the various bays along the eastern coast of Novaya Zemlya (with a further 4824 containers dumped in the Novaya Zemlya trough) with a total activity of 1230 TBq<sup>58</sup>.

The dumping of these containers along the Eastern coast of Novaya Zemlya has led to elevated levels of radionuclides in bottom water and sediments at some sites in the proximity of leaking containers, but with no impacts for the wider Kara Sea, and there have been no reports of any releases from the dumped reactors containing SNF<sup>61,247–249</sup>. Before dumping, the reactor units were filled with furfural-based polymers that are designed to prevent ingress of seawater for a period of up to 500 years<sup>59</sup>. Russia had announced that they intend to raise the dumped reactors containing SNF, including those within the dumped nuclear submarine K-27, by 2030<sup>71</sup>.



**Box Fig. 3** | Locations and overall activities (TBq) of different types of waste dumped in the Atlantic Ocean, Pacific Ocean, and Arctic. Reactors without spent nuclear fuel dumped in the Atlantic and Pacific Oceans were, respectively, the Seawolf submarine reactor shell dumped by the US in 1959 and the two reactors from submarine No. 143 dumped by the former USSR in 1971. Activities stated for low-level solid waste include some intermediate-level waste dumped by the US (Atlantic and Pacific Oceans) and the former Soviet Union (Pacific Ocean and Arctic). Activities stated for low-level liquid waste represents waste that was dumped directly into the marine environment. Figure adapted with permission from the IAEA<sup>57</sup>.

#### Box 5 | What kind of accidents and losses of radioactive material have happened?

The destruction of four nuclear weapons when a US B52 plane crashed on sea ice near the Thule Air Base (now known as Pituffik Space Base) in northwest Greenland in 1968, led to the contamination of sediments within Bylot Sound with plutonium-239,240 (<sup>239,240</sup>Pu)<sup>250</sup>. Despite extensive clean-up operations, it has been estimated that 9.6 TBq of <sup>239,240</sup>Pu remain in sediments where the crash occurred<sup>251</sup>, and that levels of <sup>239,240</sup>Pu in bottom water at the crash site were 3–6 times higher than the background level due to the resuspension of contaminated sediments<sup>252</sup>.

Six of the eight known nuclear submarines that sank at sea because of accidents (i.e., not including K-27, which was dumped) remain on the seafloor<sup>57</sup> (Supplementary Table 12). Of these, the USS Thresher, USS Scorpion, K-8, and K-219 lie at depths (>2500 m) considerably deeper than their likely design crush depths. However, environmental investigations at the USS Thresher and USS Scorpion wreck sites concluded that there had been no discernible effect on the ambient levels of radioactivity<sup>253</sup>. Of the remaining two submarines, releases of

radionuclides have only been observed from Komsomolets (K-278), which lies in the Norwegian Sea at a depth of 1700 m. The estimated reactor inventory in 2020 would have decreased to ~3 PBq, but with a further 16 TBg of plutonium in the two nuclear warheads housed within the torpedo compartment<sup>67,69</sup>. Releases from the submarine's reactor were detected shortly after it sank in 1989 and as recently as 201967-69. However, monitoring around Komsomolets has shown that the releases have no impact on the overall levels of radionuclides in the Norwegian Sea due to the rapid dilution of the releases in the surrounding water<sup>70,254</sup>. K-159 sank outside of Murmansk Fjord in 2003 whilst being towed for final decommissioning and now lies at a depth of 246 m<sup>66</sup>. K-159 contained two nuclear reactors that had been shutdown since 1989 but still contained ~800 kg of SNF and an estimated total activity of some 3.4 PBg at the time of sinking<sup>255</sup>. A joint Norwegian-Russian investigation in 2014 detected no signs of any releases from the two reactors onboard K-159<sup>66</sup>. K-159 has been included in the list of objects containing SNF that Russia plans to raise from the seafloor in the Arctic by 2030<sup>71</sup>.

during the winter months<sup>62–64</sup>. However, a more recent field study of Stepovogo Bay concluded that the underwater sill between the inner and outer parts of the bay likely prevented the flushing of bottom water in the inner part of the bay where solid radioactive waste has been dumped and that this bottom water is probably only renewed through winter convection<sup>65</sup>.

### Accidents and losses involving radioactive material in the marine environment

Since the dawn of the nuclear age in the 1940s, accidents involving military aircraft, military and civilian vessels, the re-entry of satellites and spacecraft as well as the loss of industrial sealed sources have led in some situations to the release of radionuclides into the marine environment<sup>57</sup> (Box. 5). In some cases, such accidents have led to the loss of nuclear weapons and or nuclear reactors.

# Climate change impacts on objects containing radioactive material sunken or accidentally lost at sea

For a discussion of CC impacts on radioactive material from accidents and losses that have already been released into the marine environment (e.g., the accident at Thule (see Box 5)), refer to the relevant section on CC impacts on secondary sources. In other situations, it is difficult to see that the observed and projected CC impacts would have any notable impact either where releases of radionuclides to the marine environment have yet to be observed (e.g., K-15966) or in the case of Komsomolets where releases from the reactor have been ongoing for over 30 years<sup>67-70</sup> (see Box 5). Nevertheless, as mentioned for dumped waste, the issue of corrosion rates may need to be considered in these situations. In the case of K-159, the main concern would be the integrity of the remaining barriers preventing the ingress of seawater into the reactor core, although this issue will be moot if K-159 is raised as planned by 203071. For Komsomolets, changes in bottom water properties might influence leaching rates of radionuclides from the fuel matrices as well as the dissolution of the SNF itself and the corrosion of any materials maintaining the integrity of the fuel assemblies with the reactor core.

#### Climate change impacts secondary sources arising from past human activities

#### Historic contamination from nuclear weapon tests

Over 2000 nuclear detonations have been carried out, with the vast majority taking place between 1945 and the adoption of the United Nations Comprehensive Test Ban Treaty in 1996<sup>72</sup>. These include nuclear weapon tests, wartime detonations, and so-called peaceful nuclear explosions used primarily by the former Soviet Union for purposes such as construction and

extinguishing gas well fires<sup>73</sup>. In addition to direct inputs of global fallout from atmospheric nuclear weapon tests in the 1950s, 60 s, and 70 s into the oceans, further inputs to the marine environment have occurred through terrestrial runoff<sup>74</sup>. In terms of secondary sources to the marine environment, it is important to note that a number of nuclear weapon tests were carried out at coastal locations (Box. 6).

### Historic contamination from nuclear accidents and nuclear facilities

The nuclear accidents at Chornobyl and Fukushima have led to the direct contamination of the marine environment as well as the terrestrial environment, which has then led to secondary sources to the marine environment through rivers, surface runoff and, where relevant, submarine groundwater discharge<sup>75,76</sup>. When such accidents occur, the fluxes of radionuclides through these pathways are generally highest in the years following the accident and then decrease over time as the mobile fraction of deposited radionuclides is reduced through washout and physical decay<sup>77–81</sup>. Accidents and/or previous working practices at other nuclear facilities have also resulted in contamination of the terrestrial environment that has and continues to provide inputs to the marine environment via river runoff (Box. 7).

# Historic contamination from previous discharges from nuclear reprocessing facilities

The lifetime of operations at the Sellafield and la Hague nuclear reprocessing facilities have led to considerable inputs of various radionuclides into the marine environment in the past when authorized discharges were often higher than at present<sup>82,83</sup>. A fraction of the radionuclides discharged by these facilities into the Irish Sea and English Channel have accumulated in sediments around the discharge points<sup>84–88</sup>, which now act as secondary sources where radionuclides are released back to the water column over time<sup>89–91</sup>. The main controlling factors that have influenced the degree of this accumulation are the chemical behavior of the individual radionuclides discharged, the amount discharged, and the sedimentology and hydrology of the local marine areas (Box. 8).

#### Historic contamination from previous discharges from nonnuclear industries

Previous operational releases from the phosphate industry have also resulted in the considerable accumulation of naturally occurring radionuclides in the areas affected by these discharges. For example, operation releases of phosphogypsum waste and acid mine drainage from sulfide

#### Box 6 | Where did nuclear weapon tests occur in the marine environment?

Nuclear weapon tests were carried out by the US, the former Soviet Union, France, and the United Kingdom at a number of coastal locations within the Pacific and Indian Oceans as well as in the Barents Sea (Box Fig. 4). These tests have resulted in varying degrees of contamination on land and in the local marine environment<sup>256-261</sup>. In the case of Bikini Atoll,

the US Pacific proving grounds, this has resulted in prolonged restrictions on resettlement<sup>262,263</sup>. The tests carried out by the former Soviet Union in Chernaya Bay, at the southern tip of Novaya Zemlya, contaminated sediments at the site with elevated levels of radionuclides, including an estimated 11 TBq of <sup>239,240</sup>Pu<sup>264</sup>.



**Box Fig. 4** | Location and number of nuclear tests carried out at coastal locations. These nuclear tests were mainly atmospheric detonations, but underwater tests were carried out at Bikini Atoll, Enewetak Atoll, and Chernaya Bay<sup>258,261,265-270</sup>. The US also carried out three tests in the South Atlantic (high altitude) and three tests in the Pacific (one atmospheric and two underwater) at locations remote from any land mass. Contamination of Johnston Island with plutonium isotopes also occurred from the non-nuclear destruction of three missiles<sup>256</sup>.

mines from the Tinto and Odiel rivers in Spain have led to the accumulation of high levels of naturally occurring radionuclides in the river sediments and in the Huelva estuary due to pH gradients at the estuarine mixing zone<sup>92-94</sup>. A recent study of the coastal area around the phosphate production facility at Jorf Lasfar in Morocco, which began production in 1984, revealed substantial radioactive contamination in the marine environment but that such impacts were limited to an area of 10 km around the phosphogypsum discharge points<sup>95</sup>. The radiological impact on humans from such operational releases to the marine environment can arise from the use of contaminated sands in construction and through the consumption of seafood<sup>95</sup>. The main exposure through the consumption pathway is due to the elevated accumulation of polonium-210 (<sup>210</sup>Po; a decay product of <sup>226</sup>Ra) in seafood where such discharges occur<sup>96</sup>. Even when such discharges cease, elevated exposures from <sup>210</sup>Po in seafood can occur for decades afterwards, due to the residual contamination of the local marine area<sup>97</sup>.

Modeled dispersions of naturally occurring radionuclides from discharges of produced water in the North-East Atlantic in seawater have shown that any additional concentrations were far lower than typical lowend background levels for the region<sup>98</sup>. However, there is some evidence for the accumulation of <sup>226</sup>Ra and <sup>228</sup>Ra in sediments as a consequence of decades of produced water discharges from the offshore oil and gas industry in the North Sea and Norwegian Sea<sup>99,100</sup>.

#### Historic contamination of the cryosphere

The signatures of man-made radionuclides from nuclear weapon tests, fallout from the accidents at Chornobyl and Fukushima as well as the presence of naturally occurring radionuclides have been detected in glaciers and permafrost around the world as well as in the Greenland and Antarctic ice caps<sup>101-109</sup>. Glaciers will transport any deposited radionuclides along their surfaces to their terminus, which, in the case of tidewater glaciers, will mean direct inputs into the marine environment. Man-made and naturally occurring radionuclides that have been deposited on the surface of glaciers can be concentrated through adsorption to material known as cryoconite<sup>110-113</sup> that forms on the surface of glaciers and in meltwater as a mixture of mineral particles and microbial growth<sup>114</sup>. Although, even in the case of tidewater glaciers, the further transport of any inputs of radionuclides to the marine environment may be restricted due to the massive quantities of moraine transported and released by glaciers, which would promote the scavenging and sedimentation of released radionuclides<sup>112,115,116</sup>.

However, contaminated sediments that can be entrained during the formation of seasonal coastal sea-ice and river ice maybe transported further afield before being released back into the water column when such ice eventually melts<sup>117–119</sup>. These processes are particularly relevant for the releases of radionuclides that have occurred to the Ob and Yenisey rivers and

# Box 7 | What kinds of inputs of radionuclides to the marine environment have occurred through historic contamination from nuclear accidents and nuclear facilities?

The estimate for the input of <sup>137</sup>Cs to the marine environment from the Chornobyl accident in 1986 to the marine environment (16 PBq) includes contributions from terrestrial runoff at that time<sup>74</sup>, yet Chornobyl runoff has continued to influence the levels of radionuclides such as <sup>137</sup>Cs in coastal sediments and seawater<sup>229,271–274</sup>. In the case of Fukushima, direct inputs of <sup>137</sup>Cs to the Pacific Ocean have been estimated at 8.1–17.1 PBq<sup>220</sup>, but with reported ongoing inputs of <sup>137</sup>Cs to the marine environment in river runoff of 10–12 TBq/yr and 15 to 20 TBq/yr in submarine groundwater discharge<sup>76</sup>. A further secondary source of <sup>137</sup>Cs has been identified under beach sediments along the coast from Fukushima and linked to the deposition of elevated levels of <sup>137</sup>Cs are slowly being released back into the ocean at a rate of 0.6 TBq/yr<sup>131</sup>.

It has been estimated that some 63,000 PBq of radioactive contamination has been released into the West Siberian Basin, which ultimately drains into the Kara Sea due to the former Soviet Union's nuclear program<sup>275</sup>. Most of the contamination that has been released to the Ob and Yenisey rivers has occurred through accidents, emissions, and discharges from three facilities: Mayak Production Association (Mayak PA), the Mining and Chemical Industrial Combine (MCIC), and the Siberian Chemical Combine (SCC)<sup>235,275,276</sup>. Releases from these nuclear facilities have led to substantial inputs to the Kara Sea and contamination of the estuaries, river sediments, flood plains, and the surrounding watersheds<sup>212,277-280</sup>. Of note, the 1957 Kyshtym accident at Mayak PA led to the release of 740 PBq of medium- and long-lived radionuclides into the atmosphere that were deposited in a narrow radioactive plume for over 300 km<sup>281</sup>. It has been estimated that 106 PBq of intermediate-level waste was discharged from Mayak PA, directly into a tributary of the Ob River during the years 1949 to 1956<sup>235</sup>. At the MCIC on the bank of the Yenisey, an estimated 26,000 to 37,000 PBq have been into the ground within the site boundary to a depth of 190 to 475 m<sup>282</sup>, while routine discharges have been estimated to have contributed 30 to 100 TBq of <sup>137</sup>Cs into the Kara Sea<sup>276</sup>. Discharges from the SCC on another tributary of the Ob River have been estimated at 1.15 PBq, but only a fraction of these (0.4%) were radionuclides with physical half-lives longer than one year<sup>283</sup>.

As part of the US nuclear weapons program, operational releases of radionuclides over the lifetime of the Hanford site in Washington occurred to the Columbia River, which drains into the Pacific Ocean<sup>284</sup>. Con-taminated soils and groundwater at the now decommissioned complex have been shown to still result in releases of radionuclides to the Pacific Ocean<sup>285,286</sup>.

# Box 8 | What do we know about the build-up and release of radionuclides in sediments affected by previous discharges from the nuclear reprocessing facilities at Sellafield and la Hague?

When discharged to the marine environment, particle reactive radionuclides such as  $^{239,240}$ Pu and americium-241 ( $^{241}$ Am) readily bind to sediments, whereas others such as technetium-99 ( $^{99}$ Tc) and  $^{137}$ Cs are more soluble and typically remain within the water column $^{221-224}$ .

An assessment of the overall inventories of radionuclides in the English Channel in the 1990s showed that a substantial fraction of the la Hague discharges of plutonium isotopes and cobalt-60 ( $^{60}$ Co) remain in the bottom sediments of the English Channel with maximum levels found in the vicinity of the discharge point<sup>84,85</sup>. In contrast, most of the <sup>137</sup>Cs discharges appeared to have been dispersed by seawater transport through the English Channel<sup>84,85</sup>. In the case of discharges from Sellafield, various radionuclides have accumulated in inter-tidal sediments along the neighboring coastline as well as in sub-tidal sediments in the Irish Sea in amounts of the order of 10<sup>1</sup> to 10<sup>2</sup> TBq<sup>86-88</sup> (Supplementary Table 13). It was estimated that the activities of <sup>239,240</sup>Pu and <sup>241</sup>Am in sub-tidal sediments in the Irish Sea accounted for 61% and 57% of the cumulative

their estuaries, as well as from dumped radioactive waste in bays along the Eastern coast of Novaya Zemyla<sup>120-122</sup>.

### Climate impacts on secondary sources in the marine environment

The expected continuous trend of increased sea level rise, in combination with an increase in extreme wave heights, will impact the erosion of coastal sediments and the salinization of coastal areas, submarine groundwater, and estuaries, particularly during extreme sea level events (e.g., storm surges) and flooding. Such impacts will likely lead to the increased resuspension and mass transport of coastal sediments at the atoll test sites in the Pacific and coastal areas impacted by historically authorized discharges and accidental contamination. In laboratory experiments to simulate the erosion of discharges of these radionuclides from Sellafield, compared to just 8% and 2% for <sup>99</sup>Tc and <sup>137</sup>Cs at the time when these studies were carried out<sup>87,88</sup>.

These sediments in the Irish Sea and the English Channel now act as secondary sources of radionuclides back to the water column through remobilization<sup>89-91</sup>. In the Irish Sea, it has been estimated that 300 TBq of <sup>137</sup>Cs was remobilized from the tidal zone around Sellafield between 1989 and 2009, with a likely current remobilization rate of over 1 TBq per year<sup>90</sup>. Due to the impact of these secondary sources, levels of <sup>137</sup>Cs and <sup>239,240</sup>Pu in seawater around Sellafield are higher than would occur from current discharges alone<sup>90</sup>. In the case of <sup>241</sup>Am, it is the in-situ ingrowth of this radionuclide from previous discharges of its parent radionuclide <sup>241</sup>Pu that is acting as a secondary source of this radionuclide. The ingrowth of <sup>241</sup>Am has been estimated to account for an additional 8 TBq per year based on data up to 2010, approximately 200 times greater than from discharges at that time<sup>90</sup>.

sediments, resuspension of <sup>239,240</sup>Pu contaminated sediments did not result in any meaningful increase in the remobilization of <sup>239,240</sup>Pu to the dissolved phase<sup>123</sup>. The degree of remobilization of any radionuclides from such historically contaminated sites will depend on a range of factors, including the specific chemistry and speciation of the individual radionuclides, the texture and mineralogy of the sediments, and the physiochemistry of the sediments and overlying waters.

Zones where the mixing of freshwater in estuaries and submarine groundwater in coastal areas with seawater can be important sinks or sources of radionuclides<sup>80,124-126</sup>. Sea-level rise may move these mixing zones<sup>127,128</sup> and, therefore, the areas of sinks or sources further inland. It has been shown through modeling that radionuclides can partition between the dissolved and particle-bound states over the course of a tidal cycle as the

salinity of water overlying estuarine sediments changed<sup>129</sup>. Furthermore, evidence indicates that strontium-90 (<sup>90</sup>Sr), <sup>137</sup>Cs, and radium isotopes can be mobilized from freshwater sediments when they are exposed to seawater<sup>125,130</sup>, while <sup>239,240</sup>Pu is removed from freshwater as it mixes with seawater<sup>124</sup>. In the case of the Huelva estuary in Spain, uranium from phosphogypsum wastes is stripped from the acid mine river water in the freshwater/seawater mixing zone at a pH range of 4-6, but is then released back into the dissolved phase at higher pH values that are found in seawater<sup>94</sup>. Any upstream shift in this pH front may lead to the remobilization of uranium that has accumulated in sediments that previously acted predominately as a sink. Similarly, inland movement of mixing zones may result in changes in the location of precipitation and remobilization fronts, such as that reported for Fukushima <sup>137</sup>Cs under beach sediments<sup>131</sup> and radon-222 (<sup>222</sup>Rn), <sup>210</sup>Pb, and <sup>210</sup>Po in groundwater<sup>126</sup>. Salinization of land in coastal areas and along the seawater reaches of estuaries and rivers is expected to increase with further sea-level rise<sup>132</sup>, which will affect the mobility of radionuclides present in such sediments and soils<sup>133-135</sup>.

There is little information on whether seawater temperatures have any direct effect on the remobilization of radionuclides, although no effect was observed on the remobilization of <sup>239,240</sup>Pu between 4 °C and 25 °C<sup>136,137</sup>. However, the expected increase in ocean temperatures by the end of the 21st century may have other impacts on the release of radionuclides from contaminated sediments. For instance, increased warming in conjunction with increased nutrient loads in estuaries can lead to increased deoxygenation<sup>138</sup>. Spatial distribution patterns in benthic species important for bioturbation will also likely change under future climate scenarios<sup>139</sup>. Bioturbation can be the main cause of vertical transport of radionuclides into sediments<sup>140,141</sup> and can offset the redox impact of anoxic sediments by allowing oxygenated water to penetrate deeper into the sediment column<sup>142</sup>. Any decrease in oxygen levels in seawater and sediments may result in redox reactions that can change the solubility and, ultimately, the bioavailability of radionuclides<sup>143,144</sup>.

Further controls on the fate of radionuclides entering the marine environment may also be affected by CC impacts as well as ocean acidification, such as changes in the availability of organic ligands<sup>145</sup> and the weathering rates of radioactive particles that have been associated with many of the past, present, planned and accidental releases of radionuclides<sup>146</sup>.

### Climate impacts on secondary sources in the terrestrial environment

Greater extremes in precipitation and drought patterns and intensification of extreme precipitation events<sup>147,148</sup>, along with increasing use of freshwater for human and industrial uses<sup>149,150</sup>, will lead to changes in runoff patterns<sup>3</sup>. Such changes will alter the rate and magnitude of washout of any radionuclides and organic and inorganic material from affected watersheds. Typhoons that brought heavy rains to Fukushima Prefecture after the accident in 2011 led to an overall increase in <sup>137</sup>Cs in river water compared to normal river conditions, and in particular, an increase in <sup>137</sup>Cs associated with terrestrial material<sup>151,152</sup>. Similar observations of increased levels and particulate-associated <sup>137</sup>Cs were reported in rainstorm-associated washouts following the Chornobyl accident<sup>153</sup>. Increased washout of radionuclides bound to terrestrial material in such scenarios can lead to increased dissolved levels in coastal waters due to the desorption of particulate-bound radionuclides<sup>152</sup>.

The combination of increased surface air temperatures and reduced precipitation is expected to increase desertification and dust storms in affected areas<sup>154–156</sup>. This may lead to increased drying and windblown erosion of soils, simultaneously removing any radionuclides in the surface layers of soil<sup>157</sup> that can then be transferred to the marine environment via the atmosphere or rivers. Detectable atmospheric levels of fallout radionuclides associated with dust storms originating in Western Africa and Central Asia have been detected at a considerable distance from their source locations<sup>158,159</sup>, with increased levels of <sup>239,240</sup>Pu associated with dust residues sampled in Japan linked to desertification in the East Asian continent<sup>160</sup>.

The potential for dust storms to transport radioactive contamination has been documented in areas contaminated after the Chornobyl accident<sup>161,162</sup>. In 1992, a dust storm increased levels of airborne radionuclides at the Chornobyl site by one to two orders of magnitude<sup>161</sup>. More recently, in 2020, it was estimated that a dust storm following a wildfire in the Chornobyl exclusion zone resuspended a total activity of 27 GBq<sup>162</sup>.

The risk of wildfires is expected to increase with increasing surface temperatures, drought, and heat waves<sup>163</sup>. The wildfire in the Chornobyl exclusion zone in 2020 has been estimated to have released up to 1.5 TBq of <sup>137</sup>Cs, 0.6 TBq of <sup>90</sup>Sr, and 0.08 TBq of <sup>239,240</sup>Pu to the atmosphere<sup>162,164</sup>. Managed burning of areas at the United States Department of Energy nuclear facility at Savannah River, producing measurable airborne levels of plutonium and naturally occurring radionuclides<sup>165</sup>. Wildfires not only add to the potential for windblown dust, but they can impact runoff by increasing the washout of radionuclides bound to particulate material in post-fire sites<sup>166</sup>.

#### Climate impacts on secondary sources in the cryosphere

In the cryosphere, the increased retreat of glaciers<sup>167</sup>, changes in glacial meltwater runoff<sup>168</sup>, snow cover<sup>169</sup>, and timing of seasonal snow melt<sup>170</sup> will have impacts on runoff in relevant watersheds. CC impacts, including effects on seasonal runoff, can have consequences for the breakup of frozen rivers and the formation of ice jams<sup>171,172</sup> that can scour riverbeds and cause flood events leading to increased transport of sediment and any associated radionuclides in rivers<sup>173-175</sup>. Due to increasing air temperatures, more frequent mid-winter build-up and breakup of ice jams can be predicted that may alter sediment dynamics in rivers with seasonal ice cover<sup>175,176</sup>. Changes in the supply, transport, and deposition of sediments in such rivers will ultimately impact the input of radionuclide contamination to the marine environment. In terms of tidewater glaciers, acting as secondary sources of radionuclides, any impact from increased glacial melt will need to be considered against any retreat of glaciers onto land. However, increases in meltwater ponds on glacial surfaces<sup>177</sup> may promote the conditions for the formation of cryoconites and the accumulation of atmospherically deposited radionuclides by such material.

Maximum ice thickness and duration on the Ob and Yenisiey rivers and other Arctic Russian rivers have decreased<sup>178</sup>, as has the period when the Kara Sea coastal zone has been ice-free due to later freeze and earlier melt timings<sup>179</sup>. Coupled with further reductions in Arctic sea-ice cover<sup>180</sup>, the capacity and distance that atmospherically deposited radionuclides and entrained contaminated river and estuarine sediments might be transported by river/sea-ice would be expected to decrease<sup>181</sup>.

Ongoing permafrost thaw and deepening of the active layer in permafrost-affected soils<sup>182,183</sup> will alter the mobility of radionuclides in affected regions depending on whether such areas become wetter or drier<sup>184</sup>. Recent studies have linked permafrost thaw to increased fluxes of global fallout tritium<sup>101</sup>, naturally occurring<sup>222</sup>Rn<sup>185,186</sup> as well as increased levels of naturally occurring<sup>228</sup>Ra in seawater in the central Arctic Ocean due to greater transport of shelf-derived sediments<sup>187</sup>.

#### Climate change impacts potential sources of radionuclides in the marine environment

As a result of various human activities, operational and legacy radioactive wastes can be found at numerous coastal locations and further inland within river catchment areas, which could potentially result in further sources of radionuclides in the marine environment in the future. Such wastes include material from military nuclear activities, phosphogypsum from the processing of phosphate rock, uranium, and other metal mine tailings, and fly ash from coal-fired power plants and other industries<sup>35,188–192</sup>. In many cases, such wastes have already led to the input of some radioactive material into the marine environment<sup>93,193,194</sup>.

#### Legacy military wastes

Nuclear weapon tests have led to varying degrees of local contamination of the test site areas. For example, the US conducted 42 near-surface and air

burst nuclear tests at Enewetak Atoll in the Marshall Islands in the Pacific Ocean between 1948 and 1958<sup>195</sup>. In the 1970s following the decision to return the atoll to the Marshallese, the US government cleaned up radioactive debris and contaminated soil, which were then disposed of in an unlined nuclear test crater on Runit Island<sup>195</sup>. The radioactive material was then covered with a 10 m high, 45 cm thick concrete dome (Runit Dome), with the dome then surrounded by a ~3 m high wall to reduce wave impact<sup>195</sup>. Runit Dome has been estimated to contain 545 GBq of transuranics (e.g., plutonium isotopes) in the waste material that was disposed, with a further 30 to 1000 GBq of activity that was already present following the nuclear test that formed the crater<sup>195,196</sup>. The radioactive waste within and below the floor of Runit Dome is known to be exposed to the migration of groundwater and intruding seawater through fractures formed in the underlying bedrock by the original test explosion<sup>195,197</sup>.

In addition to nuclear weapon testing, the pursuit of the nuclear arms race during the cold war has led to other legacy radioactive waste problems. For example, considerable solid and liquid radioactive waste has been stored at the Mayak PA site within the West Siberian Basin due to nuclear weapon production and nuclear reprocessing. This includes vast volumes of low level (429 million m<sup>3</sup>) and medium level liquid waste (2.2 million m<sup>3</sup>) that has been released to a series of industrial reservoirs and a nearby lake (Lake Karachay), with an estimated combined activity in 2010 of 4843 PBq<sup>198</sup>.

#### Operational and legacy non-nuclear industry wastes

The vast majority of the phosphogypsum waste produced in the past has been stored on land either as dry stacks or, more commonly, as wet stacks or in storage ponds<sup>35</sup>. It was estimated that by 2006, a total of 2.6–3.7 billion tonnes of phosphogypsum waste had been accumulated in over 50 countries, and it has been predicted that global phosphogypsum wastes will reach 7-8 billion tonnes by 2025<sup>35</sup>. Based on a reported mean <sup>226</sup>Ra activity concentration in phosphogypsum of 1  $Bq/g^{35}$ , this would give an estimate of 7–8 PBq for <sup>226</sup>Ra alone in waste stacks by 2025. Currently, the highest number of phosphogypsum stacks occurs in the US, China, and Morocco, with some measuring in square kilometers<sup>35,199</sup>. In some countries, tighter regulations require new stacks to be constructed using liners to prevent groundwater contamination, but even where such regulations are now in place, older stacks constructed without liners can still be present<sup>5</sup>. Leaks from both lined and unlined stacks have been reported, as well as sudden failure and draining due to the formation of sinkholes directly under the stacks<sup>35,200,201</sup>. Accidental releases from phosphogypsum stacks can have a considerable impact on coastal ecosystems, although this is usually due to the acidic nature of the released water, elevated levels of nutrients, and even other contaminants rather than through any radiological impact<sup>35,200,201</sup>.

The burning of coal produces fly ash that can be enriched in levels of naturally occurring radionuclides compared to the original coal as well as typical soils<sup>202,203</sup>. The combustion of coal in thermal power plants alone is reported to produce more than 454 million tonnes of fly ash per year globally<sup>204</sup>, with additional fly ash production from other coal-utilizing industries<sup>192,205</sup>. Fly ash that is not reused is stored in landfills, slag heaps, or slurry ponds, which can lead to the contamination of surface water as well as groundwater, particularly where such storage sites are unlined<sup>203,206,207</sup>.

#### Climate impacts on potential sources

Where operational and legacy wastes are located on coastal sites, the risk of flooding and damage to storage facilities will increase due to increased sea level rise and intensification of extreme sea level events (e.g., storm surges associated with tropical cyclones). The degree of risk will depend on the exposure of individual sites to these changes. In the case of Runit Dome, the top of the dome is only 7.6 m above mean sea level, with current sea levels already eroding its concrete edges<sup>208</sup>. The original aim of the dome was to confine and secure radioactive waste around Enewetak Atoll so as to aid the repopulation of the atoll, yet its construction was not in line with US Nuclear Regulation Commission regulations<sup>195</sup>. Currently, there are concerns that the dome may be breached (e.g., during a tropical cyclone) with the resulting release of radioactive waste into the lagoon and surrounding waters. In 1980,

an assessment concluded that the dome would withstand storm surges and typhoons and that there would be no added impact from any releases as the lagoon within the atoll was already contaminated with higher levels of transuranics from other nuclear tests<sup>209</sup>. However, the waste in Runit Dome has an areal activity concentration that is 1000 times higher than sediments in the lagoon<sup>195</sup>. Although Pacific typhoons typically form to the east of the Marshall Islands, the northern atolls, including Enewetak are more prone to typhoons than the southern atolls, with sustained storm winds of 64–119 km/h striking the area around every four to seven years on average<sup>210</sup>.

At the Mayak PA site, there is a concern that flooding or any failure of the reservoirs that have been used to store vast volumes of radioactive waste will result in massive releases of radionuclides to the Ob River system and ultimately to the Kara Sea. Average annual air temperatures in the area have increased by 1.6 °C to 2.0 °C, with an increase in annual precipitation of 32.8 mm from 1966 to 2018<sup>211</sup>. Little information is available concerning the resilience of these reservoirs to future climate scenarios and their vulnerabilities to extreme events. In 1967, parts of Lake Karachay at Mayak PA dried out, with ~20 TBq of mainly <sup>90</sup>Sr and <sup>137</sup>Cs resuspended from lake sediments by a tornado that contaminated land in the area<sup>212</sup>. In this case, mitigation was carried out in 2015 by covering the lakebed with rocks to prevent any atmospheric resuspension of contaminated sediments should the lake dry out again<sup>212</sup>.

For the vast number of phosphogypsum stacks, CC impacts on potential releases will typically depend on whether the waste is stored as dry or wet stacks. In the case of wet stacks, the main issue is the capability of the stacks to cope with heavy precipitation events coupled with increases in the intensity of tropical cyclones, where relevant. Such concerns are heightened for sites that no longer support active mining and may have aging infrastructure<sup>201</sup>. In the Gulf Coast of the US, heavy precipitation events in 2004, 2005, and 2012 led to spills from breached phosphogypsum stack walls<sup>213,214</sup>. For dry stacks exposed to extreme heat events, airborne dust can be a problem if the surface of the phosphogypsum waste completely dries out<sup>35</sup>. In addition, the formation of any cracks within the stack through drying out can allow greater entry of subsequent rainfall that can leach radionuclides and destabilize the accumulated waste<sup>35</sup>. The continued need for phosphate fertilizer and feed, as well as tighter regulations on direct discharges of phosphogypsum waste, will inevitably lead to more waste stacks in production countries, which will need to withstand regionally relevant CC impacts.

Similarly, it can be expected that fly ash waste sites and mine tailings will increasingly be exposed to heavy precipitation events and extreme heat events that may lead to increased leaching, spills, atmospheric inputs, and even the risk of the collapse of solid waste piles into watercourses<sup>215-219</sup>.

#### Conclusion

The likely effects of CC impacts and ocean acidification on existing sources of radionuclides to the marine environment are summarized in Supplementary Table 14. The main overall effect on operational releases for the nuclear and non-nuclear sectors is likely to be increased interference or prevention of normal operations due to weather-related events. Although relatively minor at present on a global basis, there is the potential for more frequent and longer-lasting interruptions of normal operations within these sectors towards the end of the 21st century. Increases in operational releases will be associated with any net gain in the number of discharging facilities (i.e., any increase in facilities via new build versus any decrease via the decommissioning of existing NPPs) and/or increased production. However, such effects may be offset by future mitigation and adaptation to improve operational resilience against increasing CC impacts and further regulation and/or possible improvements in abatement technologies to reduce the current levels of radionuclides in such operational releases. For dumped waste and sunken submarines, this study raises some questions regarding CC impacts and ocean acidification on corrosion rates and the fate of any releases, for which there are currently no clear answers. In terms of effects on secondary sources, certain CC impacts (e.g., sea level rise and changes in precipitation patterns) will lead to changes in inputs of radionuclides to the marine environment. For other CC impacts, such as increasing wildfires and dust storms, the likelihood of further inputs of radionuclides to the marine environment may increase. However, based on the known current levels of radionuclides in secondary sources in the marine and terrestrial environments as well as the cryosphere, CC driven changes in inputs may be measurable, but not necessarily of concern in terms of radiological impact. The occurrence and magnitude of any changes in input will depend on the radionuclide, initial environmental conditions, and the inter-play between the various local and or regional-specific CC drivers and responses as well as any unexpected feedback impacts. Although outside the scope of this review, it should be recognized that CC impacts and ocean acidification are also likely to result in effects on speciation, bioavailability, uptake, and fate of radionuclides in the marine environment. Likewise, the exposure of marine organisms to radioactive contamination may alter through CC-forced migration of species and phenological changes in life histories. Ultimately, the impacts on marine organisms should be evaluated employing holistic approaches, considering the emerging interactive impacts of multiple CC drivers, radionuclides, and other co-occurring contaminants.

For the effects of CC impacts on releases from potential sources of radionuclides to the marine environment (summarized in Supplementary Table 15), there is scope in certain situations for unplanned releases of radionuclides and other hazardous wastes that may have radiological impacts on local to regional scales. Understanding the risks associated with the relevant CC impacts and the likelihood of any unplanned releases occurring from such potential sources and their radiological impact will be important when considering the need for any future mitigation and adaptation approaches.

In conclusion, it is clear that there is a need to understand and assess the individual and combined effects of local or regionally relevant CC impacts on existing and potential sources of radionuclides to the marine environment to meet environmental and management challenges under future climate scenarios.

#### Data availability

No new analytical data were generated or presented in this review. However, estimates for global discharges of groups of radionuclides to the marine environment from nuclear power plants, the offshore oil and gas industry, and phosphogypsum wastes were made based on already available data. Information on the approaches used to derive these estimates is given in the footnotes to the respective tables in the Supplementary Information. Further, a brief description of the approach used to identify information included in this review is given as Supplementary Methods. Supporting data and information is given in Supplementary Tables 1 to 15. All sources of data, including databases where data was extracted, are stated. Derived averages for discharges from different reactor types are given in Supplementary Data 1. Data for activity discharged, produced water discharged and reinjected, and production data for Norway and the UK in 2020 are given in Supplementary Data 2.

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#### **Author contributions**

J.P.G. led the writing and coordinated the development of the manuscript. V.H., N.C., M.S., and I.O. provided input to the development and editing of the manuscript. V.H. developed Fig. 1, and N.C. developed Box Figs. 2–4.

#### **Competing interests**

The authors declare no competing interest.

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