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Sea shells record large biases from the marine bomb-14C curve in NW European seawater between the late 1960s and 2019

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The Northeast Atlantic alone has received 1.2 PBq of ^{14}C as liquid and gaseous releases from European nuclear fuel reprocessing plants (NRPs) between the 1950s and present. The input of reprocessing-^{14}C has the potential to elevate the regional ^{14}C content of seawater, sediments and marine biota above the ambient levels expected from the bomb-^{14}C. Yet, a comprehensive assessment of the time evolution of $F^{14}C$ in seawater is still missing for the Northwestern European Seas. Moreover, the least-well studied period of time (1990's onward) corresponds to the largest liquid ^{14}C releases reported by the Sellafield and La Hague NRPs. In this study, we aim at better constraining the temporal changes of $F^{14}C$ between the late 1960s and 2019, and to delimit the area of influence of reprocessing discharges with regard to ^{14}C. To this end, we combine Accelerator Mass Spectrometry techniques and a novel archive of bivalve shells that inhabited the Irish Sea, the North Sea, Norway and the Bay of Biscay throughout the main period of reprocessing-^{14}C discharge. The shells are made of aragonite, and thus, they can be used as an analogue of the past seawater $F^{14}C$. The shell-based $F^{14}C$ data can be accurately placed in the temporal context because the animals have a known capture date and short lifespan of two years. The reconstructed $F^{14}C$ values vary between ~1 and ~3 after the 1970s. This range of $F^{14}C$ values is even larger than the one displayed by the atmospheric bomb peak (1 - 1.9). To investigate if the excess ^{14}C is related to the reprocessing releases, we use a simple box model that simulates the seawater $F^{14}C$ by mixing bomb and reprocessing-^{14}C, as well as the naturally occurring $^{12,14}C$. In shells from the southern North Sea, the $F^{14}C$ increases 0.1-0.4 above ambient levels after the mid-1990s in response to increased discharge rates of liquid ^{14}C from the La Hague plant. Similarly, the shells collected in the Irish Sea show two consecutive peaks in the mid-1990s ($F^{14}C$ ~ 2.0) and 2000s ($F^{14}C$ ~ 2.2) that can be attributed to peak discharge rates of liquid ^{14}C reported by Sellafield. The $F^{14}C$ in shells from the eastern coast of the UK and Norway are within the range of the ambient values, which indicates the expected rapid dilution of the reprocessing signal with open ocean waters. In previous studies, the bomb-^{14}C marine curve has been used as a benchmark, among others, to estimate the age and growth rate of calcifying animals, to date marine sediments, and to investigate water mass mixing and circulation timescales. Given the biases from the marine bomb-^{14}C curve unraveled by the shell data, we suggest that liquid

releases from the NRPs should not be disregarded when applying ^{14}C as a chronological or circulation tool to marine samples collected in the Irish Sea and parts of the North Sea over the last 5 decades.