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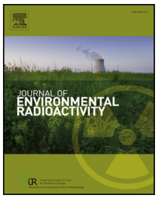

Relative comparison of tissue specific bioaccumulation and radiation dose estimation in marine and freshwater bivalve molluscs following exposure to phosphorus-32

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ABSTRACT

With respect to environmental protection, understanding radionuclide bioconcentration is necessary to relate exposure to radiation dose and hence to biological responses. Few studies are available on tissue specific accumulation of short-lived radionuclides in aquatic invertebrates. Short-lived radionuclides such as ^{32}Phosphorus (^{32}P), although occurring in small quantities in the environment, are capable of concentrating in the biota, especially if they are chronically exposed. In this study, we firstly compared tissue specific bioaccumulation and release (depuration) of ^{32}P in adult marine (*Mytilus galloprovincialis,* MG) and freshwater bivalve molluscs (*Dreissena polymorpha,* DP). Secondly, using the Environmental Risk from Ionising Contaminants Assessment and Management (ERICA) tool, we calculated tissue specific doses following determination of radionuclide concentration. Marine and freshwater bivalves were exposed for 10 days to varying ^{32}P concentrations to acquire desired whole body average dose rates of 0.10, 1.0 and 10 mGy d^{-1}. Dose rates encompass a screening dose rate value of 10 μGy h^{-1} (0.24 mGy d^{-1}), in accordance with the ERICA tool. This study is the first to relate tissue specific uptake and release (via excretion) of ^{32}P from two anatomically similar bivalve species. Results showed highly tissue specific accumulation of this radionuclide and similarity of accumulation pattern between the two species. Our data, which highlights preferential ^{32}P accumulation in specific tissues such as digestive gland, demonstrates that in some cases, tissue-specific dose rates may be required to fully evaluate the potential effects of radiation exposure on non-human biota. Differential sensitivity between biological tissues could result in detrimental biological responses at levels presumed to be acceptable when adopting a 'whole-body' approach.

1. Introduction

Short lived radionuclides such as ^{32}Phosphorus (^{32}P, radiophosphorus), although occurring in small quantities in the environment, may be capable of accumulating in aquatic biota (Smith et al., 2011). This is particularly so if the radionuclide is continuously discharged in the environment, and the biota is chronically exposed. In this context, ^{32}P is discharged into aquatic systems from various sources. For example, in England and Wales, 7, 5.2 and 5.7 GBq of ^{32}P was discharged in 2015 as liquid waste from educational, medical (i.e. hospitals) and other establishments (e.g. research, manufacturing and public sector) respectively (RIFE, 2015). In terms of environmental concentrations, ^{32}P reference conditions in Scotland (i.e. concentrations that result in a total ingested dose for humans of 0.10 mSv y^{-1} if consumed at 2 L day^{-1}), are set at 57 Bq L^{-1} (DWQR, 2014), with recorded values (2005–2013) averaging 0.27 ± 0.21 Bq L^{-1} in the River Clyde (Erskine Habour, King George V Dock), Scotland (SEPA, 2013). ^{32}P was chosen due to ease of use in an experimental setting and as a surrogate for beta/gamma emitting radionuclides ^{137}Cs and ^{90}Sr. Phosphorus in the natural environment serves as an essential nutrient, and in common with non-radioactive counterpart, radioactive phosphorus (^{32}P) would have similar exposure pathways and bioaccumulation pattern in the tissues.

In terms of human health protection, contaminated organisms could pose a risk to health via the food chain (Jha, 2004, 2008; Aoun et al., 2015; Yang et al., 2015). ^{32}P uptake in humans may occur via dietary

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Abbreviations: AM, Adductor muscle; Bq, Becquerel; CF, Concentration factor; DG, Digestive gland; DP, *Dreissena polymorpha*; ERICA, Environmental Risk from Ionising Contaminants: Assessment and Management (ERICA); IMW, Internal mussel water; IR, Ionising radiation; LSC, Liquid Scintillation Counting; ME, *Mytilus edulis*; MG, *Mytilus galloprovincialis*; mGy d^{-1}, Milligray per day; y, Year

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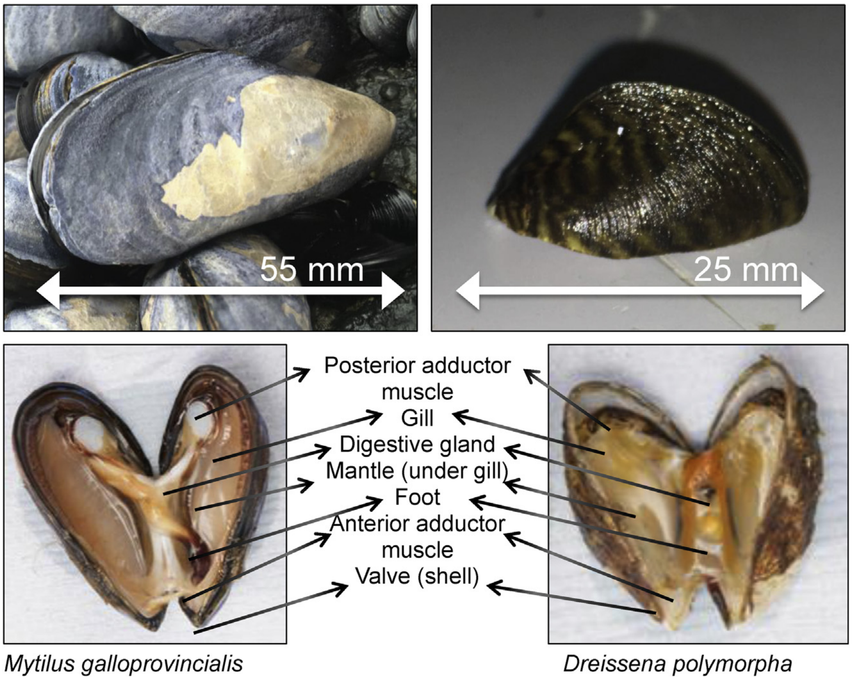

Fig. 1. Comparative external features and anatomy of *Mytilus galloprovincialis* (left) and *Dreissena polymorpha* (right).

pathways, with dose being higher in the foetus and breastfed infants, than the adult (Oatway et al., 2008). Understanding radionuclide concentration patterns in biota allows for the development of adequate protection strategies, with the aim of reducing potential human dose while maintaining environmental sustainability. Despite continuous and prolonged use in industry, and subsequent discharges, no studies to our knowledge have investigated tissue specific accumulation of ^{32}P in aquatic biota.

Bioaccumulative abilities in aquatic bivalves, an important group of invertebrates of ecological and economic importance, has been identified in scientific literature. This is notably to ubiquitous, long-lived radionuclides such as ^{134}Cs, ^{210}Po, ^{210}Pb and ^{3}H (Evans, 1984; Jha et al., 2005; Kalaycı et al., 2013; Feroz Khan et al., 2014; Dallas et al., 2016a; Metian et al., 2016; Pearson et al., 2018). However whole body accumulation and dose are often (but not always) the focus of such studies. Sufficient data are not available for tissue specific accumulation of short-lived radionuclides. It is well accepted that in common with other contaminants (Al-Subiai et al., 2011, 2012; Dallas et al., 2013; Di et al., 2011, 2017), radionuclides accumulate in the biota in a tissue specific manner. Whole-body determination of radionuclide bioaccumulation levels is important for risk assessments, however for biomonitoring and biological response studies (including sensitive transcriptomics and proteomics studies), it is important that tissue specific information is generated. Radionuclide uptake disparity amongst tissues has been highlighted in studies from Jha et al. (2005), Jaeschke et al. (2011), Dallas et al. (2016a) and Pearson et al. (2018) where tritium accumulation in bivalve (*Mytilus* sp.) tissues were observed to be highly specific. Digestive gland (hepatopancreas/gut), gill and foot showed higher concentrations following exposure to varying amounts of tritium (5–15 MBq L^{-1}). Such trends are followed in green and brown mussels (*P. perna, P. indica*), where digestive gland showed maximum ^{210}Po/^{210}Pb activity over other biological soft tissue and shell (Feroz Khan and Godwin Wesley, 2012). Furthermore, in scallop (*Pecten maximus*) soft tissue, ^{241}Am was predominantly concentrated in the mantle and digestive gland, whereas ^{134}Cs was mainly present in the adductor muscle and mantle (Metian et al., 2011). In environmental protection terms, understanding radionuclide accumulation is necessary to relate exposure, to radiation dose and to determine potential biological responses. Exposure to ionising radiations (IR) can occur via multiple aqueous and dietary pathways, the behaviour and fate of radionuclides when accumulated in specific biological tissues or organs in the aquatic biota could be influenced by many factors and may vary significantly under different exposure scenarios (Pearson et al., 2018). Given that radionuclides accumulate differentially in the tissues, from a biomonitoring perspective, whole-body bioaccumulation monitoring is therefore not necessarily sufficient in fully protecting aquatic biota from the exposure. This is particularly important as differential tissue sensitivity could result in a detrimental biological response at levels presumed to be acceptable.

Dosimetry models, such as the Environmental Risk from Ionising Contaminants Assessment and Management (ERICA) Tool have been developed to evaluate radiological risk to aquatic and terrestrial biota (Brown et al., 2008). Risk is assessed by comparing a dose rate in a reference organism to a dose rate of 10 μGy h^{-1} (0.24 mGy d^{-1}), a “screening dose rate” whereby no effect to populations of biota is expected (Garnier-Laplace and Gilbin, 2006; Garnier-Laplace et al., 2008). Though dosimetry models are of great assistance in radiobiological research, ERICA tool predicted dose rates presume homogeneous radionuclide distribution within biota, which are represented as ellipsoidal shapes (Beresford et al., 2007). In order to adequately estimate radiological risk to biota, we require a greater knowledge of tissue specific radionuclide concentrations in a range of organisms, the transfer pathways, concentration factor, dose rate and an evaluation of any possible biological effects are required. Such data may also help pinpoint key tissues of interest for biomonitoring purposes.

The presence of radionuclides is of concern for both marine and freshwater environments. The marine species *Mytilus galloprovincialis* (MG) and freshwater *Dreissena polymorpha* (DP) were therefore selected in this study (Fig. 1). Although marine species might not be used to assess the risk in the freshwater environment or vice-versa, it is nevertheless important to estimate relative radionuclide accumulation in the biota belonging to same biological or taxonomic group. This would help to identify the most sensitive species for environmental protection. These two species exhibit anatomical similarities, prevalence within respective water bodies and have known ability to concentrate contaminants within tissues. They are widely distributed and extensively used for ecotoxicological studies (Bayne, 1976; Chatel et al., 2012; Dallas et al., 2012, 2013; Binelli et al., 2015; Jaeschke