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²¹⁰Po and ²¹⁰Pb in the tissues of the deep-sea hydrothermal vent mussel *Bathymodiolus azoricus* from the Menez Gwen field (Mid-Atlantic Ridge)

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ARTICLE INFO

Article history: Received 3 June 2010 Received in revised form 13 October 2010 Accepted 14 October 2010 Available online 3 December 2010

Keywords:

²¹⁰Po

²¹⁰Pb

Radionuclide Bathymodiolus Bioaccumulation Hydrothermal vent

ABSTRACT

The hydrothermal deep-sea vent fauna is naturally exposed to a highly specific environment enriched in potentially toxic species such as sulfides, metals and natural radionuclides due to the convective seawater circulation inside the oceanic crust and its interaction with basaltic or ultramafic host rocks. However, data on radionuclides in biota from such environment are very limited. An investigation was carried out on tissue partitioning of ^{210}Po and ^{210}Pb , two natural radionuclides within the ^{238}U decay chain, in Bathymodiolus azoricus specimens from the Mid-Atlantic Ridge (Menez Gwen field). These two elements showed different distributions with high ^{210}Pb levels in gills and high ^{210}Po levels in both gills and especially in the remaining parts of the body tissue (including the digestive gland). Various factors that may explain such partitioning are discussed. However, ^{210}Po levels encountered in B. azoricus were not exceptionally high, leading to weighted internal dose rate in the range 3 to 4 μGy h $^{-1}$. These levels are slightly higher than levels characterizing coastal mussels ($\sim 1~\mu\text{Gy}$ h $^{-1}$).

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1. Introduction

Hydrothermal vents provide hot, reduced and acidic fluids enriched in various compounds, such as hydrogen sulfide, methane, carbon dioxide and various metals (see review by German and Von Damm, 2007). These fluids are also enhanced in ²³⁸U decay-chain nuclides such as ²²⁶Ra, ²²²Rn, ²¹⁰Pb and ²¹⁰Po (e.g. German et al., 1991; Kadko, 1996; German and Von Damm, 2007). Hydrothermal fluid composition is very variable on both the spatial and temporal scales. The starting fluid is mainly seawater which is modified by processes occurring within the oceanic crust along the hydrothermal flow path. Among these processes, water-rock interaction and phase separation are the most important. The overall influence of biological activity on the vent-fluid geochemistry as well as the significance of magma degassing (due to volcanic eruptions and/or diking events) versus the more "steady-state" venting has yet to be established (German and Von Damm, 2007). In particular ²¹⁰Po-degassing has been reported during mid-ocean ridge and seamount volcano eruptions (e.g. Rubin, 1997).

In biota, ²¹⁰Po (half-life of 138.4 days) and its grandparent ²¹⁰Pb (22.3 years) present ratio generally greater than one (Cherry et al., 1992). Indeed, ²¹⁰Po accumulates to high levels in the tissues of various marine organisms (Heyraud and Cherry, 1979; Heyraud et al, 1988;

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Stepnowski and Skwarzec, 2000). In addition, ²¹⁰Po is an alpha-emitting radionuclide and is the main contributor to the natural internal radiation dose received by marine organisms (Cherry and Heyraud, 1982; Carvalho, 1988). The fact that polonium has been found to be correlated with sulfur-binding protein and other sulfur-seeking metals (e.g. Stewart et al., 2007) enhanced the interest of studying this element in sulfur-rich environments such as hydrothermal vents.

The peculiar and highly productive dense fauna colonizing hydrothermal vents is not dependent on photosynthesis but rely on microbial chemosynthetic primary producers using reduced chemicals present in the hydrothermal fluid (e.g. Fisher, 1990; Cavanaugh et al., 2006). The hydrothermal vent bivalve Bathymodiolus azoricus is an endemic and dominant species at various hydrothermal vent sites along the Mid-Atlantic spreading center where it can form extensive beds surrounding the active area (Desbruyères et al., 2000). The adaptive success of B. azoricus in such challenging environments is due to its mixotrophic nutrition. Indeed, this species hosts both methanotrophic and thiotrophic bacterial symbionts in its gill cells (Fiala-Médioni et al., 2002; Duperron et al., 2006). This dual symbiosis allows B. azoricus to cope with changes in geochemical regimes at the vent sites providing them a strong nutritional advantage. In addition, B. azoricus also derives its nutritional requirements by direct suspension-feeding (Riou et al., 2010).

While several studies on the presence and effects of metals on *B. azoricus* have been carried out (Geret et al., 1998; Company et al., 2008; Colaço et al., 2006; Kádár et al., 2006; Cravo et al., 2007; Cosson et al.,

2008), to the best of our knowledge no data has been reported regarding ²¹⁰Po and ²¹⁰Pb in this species. Indeed, very little data exists on ²¹⁰Po and ²¹⁰Pb content in hydrothermal fauna (Cherry et al., 1992; Boisson et al., 2001; Charmasson et al., 2009). These authors have mentioned relatively high levels for these two elements in *Alvinella* and *Paralvinella* polychaetes from the East Pacific Rise (Cherry et al., 1992; Charmasson et al., 2009) and in the *Cyclope* gasteropod and *Tellina* bivalve in the Aegean Basin in the Mediterranean Sea (Boisson et al., 2001).

In the present study we determined 210 Po and 210 Pb levels in *B. azoricus* from the Menez Gwen field (Mid-Atlantic Ridge). We first present the concentration and 210 Po/ 210 Pb activity ratio variations of these naturally-occurring radionuclides and then discuss tissue distribution, the isotopes' likely trophic route, possible detoxification processes and radiation doses arising from internal exposure to 210 Po.

2. Materials and methods

2.1. Sampling

The mussels were sampled at the Menez Gwen hydrothermal vent field (Fig. 1), which was discovered in 1994 during the DIVA cruise (Fouquet et al., 1994). This vent is a shallow hydrothermal site with a depth of 850 m along the Mid-Atlantic Ridge, southwest of the Azores Triple Junction. The site is characterized by small active hydrothermal structures venting clear with low mineral particle content. The faunal communities are dominated by large patches of the bivalve *B. azoricus* abundantly dispersed in the substratum with specimen abundance ranging from 400 to 700 individuals/m² (Colaço et al., 1998).

During the MoMARETO cruise (08/07/2006–09/06/2006), mussels were sampled using the grab of the ROV Victor6000 and stored in four acoustically retrievable cages. The cages were deployed on small vent outlets before their recovery. Mussels studied in this study come from the third cage, which was recovered in May 2007 (i.e. after 9 months) by the R.V. Arquipélago. All the samples were taken from the same area since they were within the same collecting cage. They are thus supposed to have been exposed to the same physical and chemical environment and so the range in measured concentrations should not be linked to environmental differences in these parameters.

Once the mussels were brought to the surface, they were immediately placed in cool particle-free seawater in the Azorean land-based hydrothermal vent laboratory, LabHorta (Colaço and Santos, 2003), for 3 to 4 days in order to depurate them of faeces and pseudo-faeces. The animals were in good state of health, as indicated by the color of their gills.

We selected hydrothermal mussels with almost similar size. This narrow size range was chosen in order to reduce age/size effects since allometric relationships with negative slope for both ²¹⁰Po and ²¹⁰Pb in marine organisms have been reported (e.g. Cherry and Heyraud,

1991). Organs were pooled in groups of five specimens with close shell size i.e. mean shell length (± 1 standard deviation) of 54.84 (± 1.44), 54.04 (± 0.96), 53.32 (± 1.80) and 60.52 (± 3.28) mm respectively for the A, B, C and D groups. An analysis of variance (ANOVA) revealed significant difference (P<0.001) between the means of the different groups. A multiple comparison test i.e. LSD (least significant difference) was then applied in order to determine which means differed from one another (Sokal and Rohlf, 1981). Group D is characterized by a mean significantly greater than groups A, B, and C, whose means do not differ from each other (P>0.05).

For each group of 5 individuals, gills, mantles, feet and the remaining parts of the soft tissues (including the digestive gland) were removed from the shells with sterile stainless steel razor blades and the byssus threads were discarded. These soft tissues were weighed, deep frozen ($-80\,^{\circ}\text{C}$) and lyophilized (Savant RVT400) at LabHorta. The samples were then homogenized using an agate mortar and pestle, in order to avoid any external contamination. These samples were processed to determine both ^{210}Po and ^{210}Pb content through alpha-counting following chemical purification.

In order to compare these concentrations with values encountered in coastal organisms, we performed ^{210}Po and ^{210}Pb analyses in the soft tissues of the Mediterranean coastal mussel, *Mytilus galloprovincialis*, sampled in Toulon (France). The mean size of this group was 67.25 ± 3.38 mm (n = 10) and was not statistically compared with the mean sizes of the *B. azoricus* groups A–D since it is separate species (i.e. different growth rate and metabolism).

2.2. ²¹⁰Po-²¹⁰Pb determination

Concentrations of ²¹⁰Po and ²¹⁰Pb were measured using a classical technique based on two ²¹⁰Po measurements at a several month interval to allow ²¹⁰Po in-growth from the ²¹⁰Pb present in the samples. These analyses were performed at the IRSN-STEME laboratory (Le Vésinet, France). Dried samples of known weight (0.2–1.1 g) were spiked with 209 Po (E $_{\alpha}$ = 4.9 MeV) as a tracer, in order to calculate the chemical recovery of polonium from the analyzed samples after the first chemical treatment. The samples were dissolved in a glass beaker using 150 ml of a mixture of concentrated acids, HNO₃-HCl (2/3-1/3). They were then evaporated slowly, to near dryness, on a hot plate at a temperature of about 80 °C. Then, 10 ml H₂O₂ (30%) in 20 ml of concentrated HNO₃ was added to whiten the sample residues. These were slowly dissolved with 35 ml of a mixture of concentrated HNO₃-HClO₄ (30 ml/5 ml) and again evaporated to near dryness. This step was performed 2 or 3 times until all the organic material was digested (Boisson et al, 2001). The sample was then dissolved in 20 ml of HCl (6 M) (Matthews et al., 2007) and transferred into the deposition cell together with 2 rinses of the beaker using demineralised water, in order to obtain a total

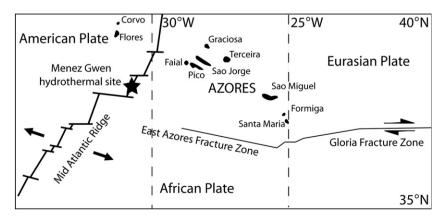


Fig. 1. Location of the Menez Gwen hydrothermal field in the Mid-Atlantic Ridge. The Azores islands and the major tectonic structures are represented. (Map adapted from Fouquet et al., 1994).

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