



Uranium accumulation in aquatic macrophytes in an uraniferous region: Relevance to natural attenuation



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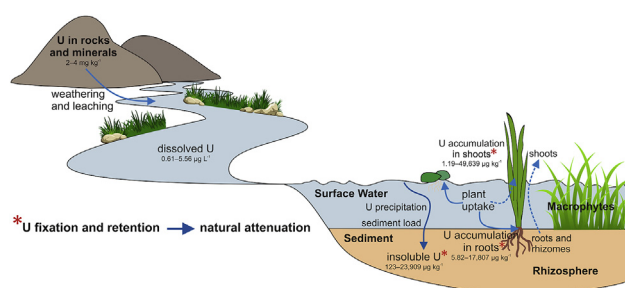
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HIGHLIGHTS

- Aquatic macrophytes showed great efficiency in accumulating uranium (U).
- Species-specific and organ-specific variations are pronounced.
- Preferential accumulation in roots contributes to U fixation in rhizosphere.
- Natural attenuation of contamination by phytostabilization in rhizosphere.
- *Rorippa sylvestris* may be considered as bioindicator for U contamination.

GRAPHICAL ABSTRACT



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ABSTRACT

Phytoremediation potential of uranium (U) was investigated by submerged, free-floating and rooted emergent native aquatic macrophytes inhabiting along the streams of Horta da Vilariça, a uraniferous geochemical region of NE Portugal. The work has been undertaken with the following objectives: (i) to relate the U concentrations in water–sediment–plant system; and (ii) to identify the potentialities of aquatic plants to remediate U-contaminated waters based on accumulation pattern. A total of 25 plant species culminating 233 samples was collected from 15 study points along with surface water and contiguous sediments. Concentrations of U showed wide range of variations both in waters (0.61–5.56 $\mu\text{g L}^{-1}$, mean value 1.98 $\mu\text{g L}^{-1}$) and sediments (124–23,910 $\mu\text{g kg}^{-1}$, mean value 3929 $\mu\text{g kg}^{-1}$) and this is also reflected in plant species examined. The plant species exhibited the ability to accumulate U several orders of magnitude higher than the surrounding water. Maximum U concentrations was recorded in the bryophyte *Scorpiurium deflexifolium* (49,639 $\mu\text{g kg}^{-1}$) followed by *Fontinalis antipyretica* (35,771 $\mu\text{g kg}^{-1}$), shoots of *Rorippa sylvestris* (33,837 $\mu\text{g kg}^{-1}$), roots of *Oenanthe crocata* (17,807 $\mu\text{g kg}^{-1}$) as well as in *Nasturtium officinale* (10,995 $\mu\text{g kg}^{-1}$). *Scorpiurium deflexifolium* displayed a high bio-concentration factor (BF) of $\sim 2.5 \times 10^4$ (mean value). The species *Fontinalis antipyretica*, *Nasturtium officinale* (roots) and *Rorippa sylvestris* (shoots) exhibited the mean BF of 1.7×10^4 , 5×10^3 and 4.8×10^3 respectively. Maximum translocation factor (TF) was very much pronounced in the rooted perennial herb

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Rorippa sylvestris showing extreme ability to transport U for the shoots and seems to be promising candidate to be used as bioindicator species.

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1. Introduction

Uranium (U) is a ubiquitous heavy metal in nature, occurring in several chemical forms in the environment. The global riverine U flux was estimated to be within the range 0.2–0.6 $\mu\text{g L}^{-1}$ as recorded by Palmer and Edmond (1993). Uranium is both a radioactive and chemotoxic naturally occurring radionuclide, and there had been great discussion on health effects associated with exposure to U and its compounds. In the natural environment, U occurs almost entirely as ^{238}U in its hexavalent state (U^{6+}), and in minor quantities as ^{235}U , and in trace quantities as ^{234}U (Reimann and Caritat, 1998). In aqueous systems U reacts with oxygen to form uranyl ion UO_2^{2+} which is highly stable and soluble, which determines its toxicity (Malczewska et al., 2003; Kalin et al., 2005; Schöner et al., 2009; Bhalara et al., 2014). Other soluble forms are UF_6 , $\text{UO}_2(\text{NO}_3)_2$, UO_2Cl_2 , UO_2F_2 , uranyl acetates, sulphates, and carbonates (Durakoviae, 1999). Several physicochemical and biological variables may influence the U speciation, bioavailability, uptake and toxicity in fresh surface waters, including pH, water hardness, natural organic matter, and microbial activity (Li et al., 2004; Srivastava et al., 2010; Wang et al., 2010; Laurette et al., 2012a,b; Markich, 2013).

The ability of aquatic plants to accumulate metals from water and/or from water–sediment interface, has been observed in several studies performed both in field or laboratory conditions (Brooks and Robinson, 1998; Cardwell et al., 2002; Fritioff and Greger, 2006; Nyquist and Greger, 2007; Rahman and Hasegawa, 2011; Pratas et al., 2014; Malar et al., 2014, 2015; Chowdhury et al., 2015; Rodriguez-Hernandez et al., 2015). Therefore, it has been generally observed that the metal concentration in aquatic plants is many folds higher than the concentration in ambient water.

The accumulation of chemical elements in aquatic plants may occur due to absorption, adsorption and/or other retention mechanisms. These physicochemical processes generally fall under the term “bioaccumulation”, when performed by living organisms. Another term, “biosorption”, is used to describe the set of mechanisms for the removal of substances from solution by biological materials (living or dead biomass), including absorption, adsorption, ion exchange, surface complexation and precipitation (Volesky and Holan, 1995; Raize et al., 2004; Gadd, 2009; Srivastava et al., 2010; Ungureanu et al., 2015). Furthermore, in aquatic environments, the chemical elements removal depends on other factors that include microbial biofilms on abiotic substrates or the growth of periphyton (Bradac et al., 2010; Stout and Nüsslein, 2010; Krawczyk-Bärsch et al., 2011; Castro et al., 2015).

In the last few decades, several aquatic plant species have been identified as accumulators of metal(loid)s and as a result they might prove useful in biomonitoring and phytoremediation techniques, namely phytodetoxification and rhizofiltration. Some examples are: *Ceratophyllum demersum* L. for Cu, Cr, Fe, Mn and Pb (Rai et al., 1995), *Eichhornia crassipes* (Mart.) Solms for Cd, Cr and As (Zhu et al., 1999; Alvarado et al., 2008), *Myriophyllum aquaticum* (Vell.) Verdc., *Ludwigia palustris* (L.) Ell. and *Mentha aquatica* L. for Cu, Fe, Hg and Zn (Kamal et al., 2004), *Lemna gibba* L. for As and U (Mkandawire and Dudel, 2005; Alvarado et al., 2008), *Potamogeton pectinatus* L. and *Potamogeton malaianus* Miq. for Cd, Pb, Mn, Zn and

Cu (Peng et al., 2008), *Callitriche lusitanica* Schotsman for As (Favas et al., 2012), *Callitriche cophocarpa* Sendtn. for Tl, Cd, Zn, Pb, Cr (Augustynowicz et al., 2014a, 2014b), *Pistia stratiotes* L (Farnese et al., 2014) and *Vallisneria natans* (Lour.) Hara (Chen et al., 2015) for As.

The ability of aquatic plants to accumulate U has also been shown in several studies, supporting their potential use in U bio-monitoring and/or in phytoremediation of U-contaminated water. Some examples are as follows: *Nymphaea violacea* Lehm. (Pettersson et al., 1993), *Zostera japonica* Ascherson & Graebner and *Z. marina* L. (Kondo et al., 2003), *Eleocharis dulcis* (Burm.f.) Trin. ex Hensch. (Overall and Parry, 2004), Algae (Kalin et al., 2005), *Lemna gibba* L. (Mkandawire and Dudel, 2005), *Phragmites australis* (Cav.) Trin. ex. Steud. and *Typha latifolia* L. (Gerth et al., 2005; Soudek et al., 2007; Carvalho et al., 2011), *Hydrilla verticillata* (L.f.) Royle (Srivastava et al., 2010), *Callitriche stagnalis* Scop. and *Fontinalis antipyretica* Hewd (Pratas et al., 2012; Favas et al., 2014).

The study area can be recognized as an important uranium deposit, where some geological prospecting studies were conducted (Pires and Vintém, 1987). However, this deposit never been target for mining, remaining a site where the U biogeochemical cycle is mediated mainly by natural processes and little subjected to impacting anthropic activities. Nevertheless, the geological context of the region seems to favor the transfer of very significant amounts of radionuclides, integrated into the radioactive decay chain of U, from the deposit to the surrounding underground waters (Pereira et al., 2010). According to these authors, the activities of the isotopes of U, radon gas and ^{226}Ra are particularly high, reaching, especially U, exceptional values for groundwaters.

The present study has been undertaken with the following objectives: (1) to explore the extent of superficial U contamination related with a non-mined deposit; (2) to determine the U concentrations in stream water, sediment and aquatic plants from this important uraniferous region; (3) to relate the U concentrations in water–sediment–plant system; and (4) to identify aquatic plants with the potential to remediate U-contaminated waters based on accumulation pattern.

2. Material and methods

2.1. Study area: location and geology

The study region is located in northeast Portugal, surrounding the village of Horta da Vilarica (HDV) (municipality of Torre de Moncorvo, Bragança district) (Fig. 1). In this region metasedimentary rocks of the Douro Group (“Schist-Greywacke Complex”), Hercynian granites and veins of quartz, aplite-pegmatite and basic rocks are present. The metasediments show large halos of contact metamorphism in the vicinity of granitic rocks (Moreira, 1985; Pires and Vintém, 1987; Dias, 2012).

The prospecting works carried out in the region have identified several sites with supergene U minerals disseminated in metasediments of the contact halos or accumulated in fractures intersecting these rocks (Moreira, 1985). In the granite, there are some quartz veins parallel to contact and, between them, numerous anastomosed lenticular veinlets of milky quartz and smoky quartz with secondary U minerals forming a stockwork-like quartz vein

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