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Radionuclide (^{226}Ra, ^{232}Th, ^{40}K) accumulation among plant species in mangrove ecosystems of Pattani Bay, Thailand

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ABSTRACT

Little is known regarding phytoremediation of radionuclides from soil; even less is known about radionuclide contamination and removal in tropical ecosystems such as mangrove forests. In mangrove forests in Pattani Bay, Thailand, 18 plant species from 17 genera were evaluated for radionuclide concentrations within selected plant parts. Two shrub species, *Avicennia marina* and *Pluchea indica*, accumulated the highest ^{232}Th (24.6 Bq kg^{-1}) and ^{40}K (220.7 Bq kg^{-1}) activity concentrations in roots, respectively. Furthermore, the aquatic species *Typha angustifolia* accumulated highest ^{232}Th, ^{40}K and ^{226}Ra activity concentrations (85.2, 363.5, 16.6 Bq kg^{-1}, respectively) with the highest transfer factors (TFs) (3.0, 2.0, 5.9, respectively) in leaves. Leaves of *T. angustifolia* had an absorbed dose rate in air (D) over the recommended value (74.8 nGy h^{-1}) that was considered sufficiently high to be of concern for human consumption.

1. Introduction

Several soil contaminants are considered urgent environmental and public health concerns. A number of inorganic contaminants occurring in soil may be taken up by plants and subsequently transferred through the food chain (Li et al., 2006b). Recent reports have documented serious health effects in certain agricultural areas of Thailand due to soil contamination by Pb and Cd poisoning, respectively (Meepun et al., 2013; Poopa et al., 2015). Contamination is also known to occur among certain mangrove forests and other wetlands in Thailand (Chaiyarat et al., 2013). In response to these events, contaminant levels in aquatic and marine ecosystems are being monitored because several wetland types act as repositories by receiving contaminants from both point and non-point sources (Palaniappan et al., 2010).

Mangrove forests are highly productive ecosystems and play an important role in coastal nutrient cycling (Ferreira et al., 2010). Mangrove sediment contains large quantities of organic matter, which influences the physicochemical properties of sediment, enhancing fine sediment deposition and increasing adsorption and trapping mechanisms of contaminants (Jennerjahn and Ittekkot, 1997; Paiva et al., 2016).

Radionuclide contamination has been documented in mangrove sediments worldwide (Carroll and Moore, 1994; Silva et al., 2006; Godiva et al., 2010). Understanding the behavior of radionuclides, including concentrations and distribution, can provide useful data for evaluating human health risk (El-Taher and Madkour, 2011). Radon, as ^{226}Ra and ^{228}Ra, is found naturally in soil and groundwater. Both nuclides occur in approximately equal proportions as aqueous forms and in concentrations <10 Bq kg^{-1} in soil and <1 Bq m^{-3} in water. Both ^{40}K and ^{232}Th are common radioactive elements in nature and occur in trace quantities; concentrations of ^{40}K in soil are approximately 400 Bq kg^{-1}, while ^{232}Th concentrations in rock are 1.6–200 Bq kg^{-1} (UNSCEAR, 2000). Concentrations depend on geological and geographical conditions as well as inputs from anthropogenic sources (i.e., fertilizer, cement) (Sabol and Weng, 1995; El-Saharty, 2013).

Pattani Bay is located in Southern Thailand. The coastal zone of Pattani Bay is used extensively for aquatic farming activities; products including marine macroalgae (*Gracilaria fisheri*), blood clam (*Anadara granosa*) and various aquatic fish are harvested for local consumption and for export overseas (Ruangchuay et al., 2007; Suwanjarat et al., 2009). Pattani Bay is also the location of mangrove forests, industries and small villages. Industrial wastewaters, mining activities and fertilizers from agricultural plantations are considered sources of radionuclide contamination in both seawater and sediment along the Pattani Bay coastline (Kaewtubtim et al., 2015).

In Pattani Bay, concentrations of radionuclides in sediments were reported to be approximately 175.5, 58.0, 252.6 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively (Kaewtubtim et al., 2015). Radium equivalent activity (Ra_{eq}) for the sediments was 277.8 Bq kg^{-1}, which is lower than the world average value of 370 Bq kg^{-1} (UNSCEAR, 2000). However,

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the external hazard index (H_{ex}) was 1.2 Bq kg^{-1}, which exceeds the recommended value of 1 Bq kg^{-1} as defined by the United Nations Scientific Committee on the Effect of Atomic Radiation (UNSCEAR) (Kaewtubtim et al., 2015). This index is used for assessing potential radiological risks to biota (UNSCEAR, 2000). In mangrove ecosystems radionuclides occurring in sediment or seawater can be transferred to biota via the food chain (Paiva et al., 2016).

Radioactive elements have been reported to cause leukemia in humans; this is particularly true for radium (Ra), which also has a long half-life (>1500 years) (Lyman et al., 1985). It is therefore essential to monitor types and concentrations of radioactive elements in the environment in order to mitigate potential adverse effects from exposure.

Phytoremediation technology employs the capabilities of green plants to take up contaminants or immobilize them in place (Sharma et al., 2015). Phytomanagement, a subset phytoremediation, can generate economic value for local populations by selecting contaminant-enriched plants to produce biofuels, bioplastics, biochar, paper and wood (Robinson et al., 2009). Two main options for phytomanagement of soils are phytoextraction and phytostabilization. Phytoextraction is defined as the use of plants to extract a contaminant from soil (the relevant plants are termed 'hyperaccumulators') (Wei et al., 2014), while phytostabilization involves the establishment of vegetation to reduce erosion of contaminated soil (plants are termed 'excluders') (Meeinkuirt et al., 2016). Extensive research has evaluated the phytoremediation potential of plants in terrestrial ecosystems contaminated by metals resulting from metal mining (Phaenark et al., 2009; Prasad et al., 2015); however, there are few reports on phytoremediation of radionuclides (De Franca et al., 2015); this deficiency is more pronounced for soils in tropical ecosystems such as mangrove forests.

The Pattani Bay coastal zone contains mangrove ecosystem plant species such as *Avicennia marina*, *Acanthus ebracteatus*, and *Eleocharis dulcis*. These are considered Pb excluders and Cd hyperaccumulators, respectively (Kaewtubtim et al., 2016). Such species may also have the capability to accumulate substantial quantities of radionuclides in radionuclide-enriched soil or sediment.

In this study we investigated the accumulation of ^{226}Ra, ^{232}Th, ^{40}K in plants for potential radionuclide phytoremediation within mangrove ecosystems along Pattani Bay in southern Thailand. The physicochemical properties of the sediment were evaluated for explaining the behavior of the radionuclides. In addition, assessment of radiation hazards are important in order to determine the rate at which radiation is received; thus, various indices for environmental risk monitoring for nuclides were assessed, such as radium equivalent activity (Ra_{eq}), external hazard index (H_{ex}), internal hazard index (H_{in}), absorbed dose rate in air (D) and annual external effective dose rate (E).

2. Materials and methods

2.1. Site description

Pattani Bay is a semi-enclosed estuarine bay measuring approximately 74 km^2, receives input from two major rivers, the Pattani and the Yaring (Suwanjarat et al., 2009; Swennen et al., 2001; Hajisamae and Yeesin, 2014). The water regime of the bay is a complex system which receives significant influences from tidal inputs and the tropical monsoon climate as well as surface runoff and drainage from the two rivers. Water depth in the bay ranges from 0.2 to 1.5 m with a maximum 5 m depth at the mouth of the bay. Cumulative annual rainfall and average annual temperature at Pattani Bay in 2014 were 2008.5 mm and 27.5 °C, respectively.

A total of five sampling sites, considered representative of mangrove ecosystems, were selected for study. The sites are located near the mouth of the bay (Fig. 1). In the study areas, dominant mangrove plants were identified and used for analysis of plant organ and for collection of rhizosphere soil.

2.2. Physicochemical properties of the mangrove sediment

Sediments from fifteen locations among the five sites were collected from the surface layer (0–20 cm) near the rhizosphere of selected plant species using a 2-cm diameter stainless steel hand corer. Samples were mixed thoroughly to create a composite sample and brought to the laboratory. Samples were oven-dried at 80 °C for 48 h. Dried samples were ground with an agate mortar and pestle and sieved to pass a 2-mm mesh plastic sieve. Samples were mixed thoroughly again and stored in plastic bags prior to physicochemical analyses.

Sediment pH was analyzed in a 1:5 suspension of sediment to deionized (DI) water using a pH meter (Accumet® AP115, USA). Organic matter (OM) was analyzed by the Walkley-Black titration method (Walkley and Black, 1934). Particle size analysis was analyzed by the hydrometer method (Allen et al., 1974). Cation exchange capacity (CEC) was analyzed by leaching with 1 N ammonia acetate buffered to pH 7.0 (Sparks, 1996). Total nitrogen (N) was analyzed by the Kjeldahl method (Black, 1965). Extractable phosphorus (Ext. P) was analyzed by the Bray II method (Bray and Kurtz, 1945). Extractable potassium (Ext. K) was analyzed by atomic absorption spectrophotometry after ammonium acetate (NH_4OAC) extraction (Sparks, 1996). ^{226}Ra, ^{232}Th and ^{40}K concentrations were determined by gamma-ray spectrometry with a high purity germanium (HPGe) detector. The energy calibration was performed using standard reference radionuclide sources ^{60}Co and ^{137}Cs, while the efficiency calibration was performed using reference samples ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K obtained from the Office of Atoms for Peace, Thailand.

2.3. Collection of plant species

Plant species from Pattani Bay mangrove forests were selected to determine radionuclide (^{226}Ra, ^{232}Th and ^{40}K) accumulation in different plant organs. The site map and specific locations are provided in Fig. 1. Individuals of each species were randomly collected for plant identification. Root samples were collected in the same area as the soil samples (0–20 cm depth). Samples were placed in a plant press and returned to the laboratory. Based on plant identification keys (Aksornkoae et al., 1992; Smitinand, 1980), species were identified and then confirmed by the Department of Forestry, Thailand.

2.4. Radionuclide analysis in plant organs

For each species under study, leaves, stems and roots were removed from three plants (i.e., three replicates) using stainless steel scissors. Samples were thoroughly washed with tap water and phosphate-free soap several times to remove any attached sediment, and finally rinsed with DI water. All plant materials were oven-dried at 80 °C for 48 h. Each sample was ground to a fine powder and sieved through a 2-mm nylon mesh sieve.

Radionuclides in the plant organs were determined using gamma-ray spectrometry with a high purity germanium (HPGe) detector of 160 cm^3 active volume with 70% relative efficiency. The absolute efficiency for each radionuclide is shown in Table 1. A p-type HPGe detector was supplied by CANBERRA (Model-GC7022). The energy calibration was performed using the standard reference radionuclide sources ^{60}Co and ^{137}Cs, while the efficiency calibration was performed using the reference samples ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K obtained from the Office of Atoms for Peace. The instrument has a resolution of 1173 and 1332 keV of ^{60}Co and 661 keV of ^{137}Cs gamma-ray line, respectively. The spectra of all samples were analyzed for ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K by GENIE-2000 spectra analysis software. The detector was enclosed in a 20 cm-thick graded lead shield (Canberra, USA) measuring 79 cm tall and 76 mm internal diameter and having a fixed bottom and moving cover to reduce external gamma-ray background.

Soil and plant samples were counted for 11 and 5 h, respectively. Prior to analysis of samples, the environmental gamma background

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