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A potential dating technique using ^{228}Th/^{228}Ra ratio for tracing the chronosequence of elemental concentrations in plants

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Abstract

We propose a radiometric method based on measurement of the radioactivity of the naturally occurring radionuclides ^{228}Ra and ^{228}Th and the derived ^{228}Th/^{228}Ra ratios in plant samples to estimate plant age and the corresponding nutritional conditions in a field-growing fern, *Dicranopteris linearis*. Plant age (tissue age) was associated with the ^{228}Th/^{228}Ra ratio in fronds, which implies the accumulation time of immobile elements in the plant tissue or the life span of the fronds. Results indicated that the accumulation of alkaline earth elements in *D. linearis* is relatively constant with increased age, while the *K* concentration is reversed with age because of translocation among plant tissues. Estimation of dating uncertainty based on measurement conditions revealed that the radiometric technique can be applied to trace chronosequential changes of elemental concentrations and environmental pollutants in plants with ages of less than 10–15 years.

Keywords: Soil-to-plant transfer; Plant age; Radium; Radioactivity measurement; Gamma-ray spectrometry; *Dicranopteris linearis*

1. Introduction

The uptake of essential elements and environmental pollutants by plants is subjected to the following factors, among others: plant species, soil properties, chemical behavior and plant age. Practically, these factors can be combined and represented as the soil-to-plant transfer factor (TF), by which the movement of an element from the nonliving to the living compartments of the biosphere is described (Chojnacka et al., 2005; Kabata-Pendias, 2004; Sheppard, 1985; Wenzel and Jockwer, 1999). Usually, the TF is widely varied among plants of the same species, even those growing at the same site. Moreover, the influence of plant age on the TF might be quite unpredictable, and the chronosequence variation cannot be predicted properly. Accumulation of an element, or a pollutant, with time in plants can be easily clarified by a hydroponic or pot experiment with designed treatments and period (Kumar et al., 1995; Salt et al., 1997, 2004; Zhu et al., 2002). However, monitoring the chronosequential uptake or translocation of elements in wild plants is difficult because of the uncertainty about their ages and the abovementioned factors. To monitor the change in elements at different plant growth stages, some studies separated plant samples into classes of different ages. Those studies relied on field observation and seasonal sampling and sometimes required methodologies with morphological or anatomical techniques to define age (Wyttenbach et al., 1995a; Goor and Thiry, 2004).

Two main chains of radioactive elements exist in the Earth's crust: the thorium (Th) series and the uranium (U) series, which originate from ^{232}Th and ^{238}U, respectively (Cowart and Burnett, 1994; Kathren, 1984). In the two long-lived series, decay cascades produce radioactive daughter nuclides, ultimately resulting in the stable isotopes of ^{208}Pb and ^{206}Pb. The behavior and distribution of these decay series radionuclides in the environment are based on their biogeochemistry and half-life ($t_{1/2}$), and the nature of their surroundings. Radionuclides at the two

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radioactive series behave differently in translocation into plants, because plants take up elements selectively. In general, for most vegetables, the soil-to-plant transfer of these natural elements/nuclides occurs approximately as Po/Pb > Ra > U > Th (Linsalata, 1994). In soil, the radium (Ra) isotopes ^{228}Ra ($t_{1/2} = 5.75$ y) and ^{226}Ra ($t_{1/2} = 1.60 \times 10^3$ y), originating from the Th and U series, respectively, are in relatively soluble chemical forms and are readily taken up by plants. This process leads to radioactive disequilibrium in plants, which could be used to determine plant age during growth, as well as turnover rate.

Measuring the radioactivity of naturally occurring radionuclides in the two chains has been widely used for determining chronology in earth and environmental sciences (Ivanovick and Harmon, 1992) but has rarely been used in plant dating and related studies, even though some plants can utilize certain amounts of natural radionuclides (Linsalata et al., 1989; Mortvedt, 1994). The ^{228}Th/^{228}Ra dating method, first proposed by Kobashi and Tominaga (1985) to date plant samples, was based on the assumption that Ra is predominantly utilized by plants all-at-once in the growing season and behaves as an immobile element in plant tissues. However, the age of the growing plants cannot be determined properly on such an assumption because most plants take up elements continuously throughout their life spans.

The fern *Dicranoptesis linearis*, widely growing in East Asia and Africa (DeVol and Shieh, 1994), is capable of concentrating Ra isotopes tens to hundreds of times higher in the normal radiation background than ordinary plants (Chao et al., 2006). In this study, we propose the use of the ^{228}Th/^{228}Ra ratio and accumulation time of ^{228}Ra to estimate the life span, or tissue age, of fronds during growth. More emphasis is placed on changes of elemental concentrations with time. We profile the concentrations of essential and trace elements, such as alkaline earth elements and potassium (K), throughout the life span of *D. linearis* and algebraically describe nutrition conditions at different growth stages. For practical purposes, we derive dating uncertainties with respect to the content of Ra in plants, tissue age, and measurement conditions.

2. Materials and methods

2.1. ^{228}Th/^{228}Ra method

Both Th and U series produce isotopes of radium, ^{228}Ra ($t_{1/2} = 5.75$ y) and ^{226}Ra ($t_{1/2} = 1.60 \times 10^3$ y), which are soluble and can readily form compounds that can be taken up by plants. Once ^{228}Ra is taken up and enters a plant tissue, such as a frond, it decays exponentially with time to ^{228}Th, which has a half-life of 1.91 years, and in turn decays through a series of alpha emissions. A schematic diagram is illustrated in Fig. 1 to describe the changes of ^{228}Ra and ^{228}Th with time in a plant tissue, in which the immobile elements are accumulated in the growing period.

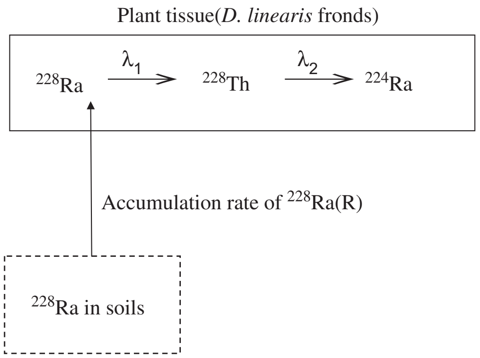

Fig. 1. Schematic diagram of the changes of ^{228}Ra and ^{228}Th in a plant tissue (*D. linearis* fronds) with accumulation time, or age.

For field-growing *D. linearis*, ^{228}Ra is expected to be continuously accumulated at a constant rate (R, in units of becquerels per kilogram per year in plant tissue such as fronds). This linear relationship will be verified from the experimental approach and will be discussed in the following sections. The accumulation and subsequent decay of ^{228}Ra in *D. linearis* tissues with time (t) in terms of atom number (N_1) is expressed as

$$\frac{dN_1}{dt} = \frac{R}{\lambda_1} - \lambda_1 \times N_1, \tag{1}$$

where λ_1 is the decay constant of ^{228}Ra, 0.121 y^{-1} (Reus and Westmeier, 1983). The plant tissues, or fronds, beginning with the blooming of sprouts, are assumed to contain no ^{228}Ra in the initial growing condition. Solving Eq. (1) gives

$$N_1(t) = \frac{R}{\lambda_1^2}(1 - e^{-\lambda_1 t}) \tag{2}$$

or

$$A_1(t) = \frac{R}{\lambda_1}(1 - e^{-\lambda_1 t}), \tag{3}$$

where $A_1(t) = \lambda_1 \times N_1(t)$ represents the radioactivity of ^{228}Ra (Bq kg^{-1}).

Thorium in the liquid phase of soils, described by distribution coefficient (k_d), is relatively insoluble (Sheppard, 1985). The amounts of ^{228}Th both directly taken up from soil and produced as a decay product of ^{232}Th inside plants are negligible relative to those of ^{228}Ra (Linsalata, 1994; Hill, 1962; Osburn, 1965). Therefore, almost all ^{228}Th (N$_2$) is formed through the decay of ^{228}Ra inside plants, expressed as

$$\frac{dN_2}{dt} = \lambda_1 \times N_1 - \lambda_2 \times N_2 \tag{4}$$

or

$$\frac{dN_2}{dt} = \frac{R}{\lambda_1} \times (1 - e^{-\lambda_1 t}) - \lambda_2 \times N_2, \tag{5}$$

where λ_2 is the decay constant of ^{228}Th, 0.363 y^{-1}.