

# Uranium mobility across annual growth rings in three deciduous tree species

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## ABSTRACT

Black walnut (*Juglans nigra*), slippery elm (*Ulmus rubra*), and white ash (*Fraxinus americana*) trees were evaluated as potential archives of past uranium (U) contamination. Like other metals, U mobility in annual growth rings of trees is dependent on the tree species. Uranium concentrations and isotopic compositions (masses 234, 235, 236, and 238) were analyzed by thermal ionization mass spectrometry to test the efficacy of using tree rings to retroactively monitor U pollution from the FFMPC, a U purification facility operating from 1951 to 1989. This study found non-natural U (depleted U and detectable <sup>236</sup>U) in growth rings of all three tree species that predated the start of operations at FFMPC and compositional trends that did not correspond with known contamination events. Therefore, the annual growth rings of these tree species cannot be used to reliably monitor the chronology of U contamination.

## 1. Introduction

Between 1951 and 1989, the Fernald Feed Materials Production Center (FFMPC), located in southwest Ohio (Fig. 1a), produced high purity U metal and relatively minor amounts of thorium for the United States nuclear industry. An estimated 54% of this U was from natural ores; however, in 1962, the FFMPC began to process recycled U (20% was enriched U and 26% was depleted U; Voilleque et al., 1995). This was a 4.25 km<sup>2</sup> site that had previously been used as agricultural land (DOE, 2011). Nearly continuous releases of U were documented to have occurred in the form of solid spills, aqueous U, particulate matter, and aerosols. During production years, it is estimated that 90–450 Mg of U metal was released into the environment, ultimately contaminating the Great Miami Aquifer (Fig. 1; Morris et al., 1996; Sidle and Lee, 1996; FEMP, 1998). In 1989, the facility was declared a Superfund Site. Remediation began in 1991 with contaminated soils and other materials being moved off site or into an on-site disposal facility, and continues with groundwater treatment. Today, the site is a nature preserve. The FFMPC is therefore representative of a site affected by nuclear contamination that has undergone a complete life cycle (i.e. production, contamination, remediation). It is a fitting locality for testing various methods of detecting historical U pollution.

Recent studies suggest that non-natural, nuclear contamination persists in the region surrounding the FFMPC (Conte and Widom, 2013; Tortorello et al., 2013; Widom and Kuentz, 2010a). Studies of tree bark compositions in the vicinity of FFMPC provide a time averaged record

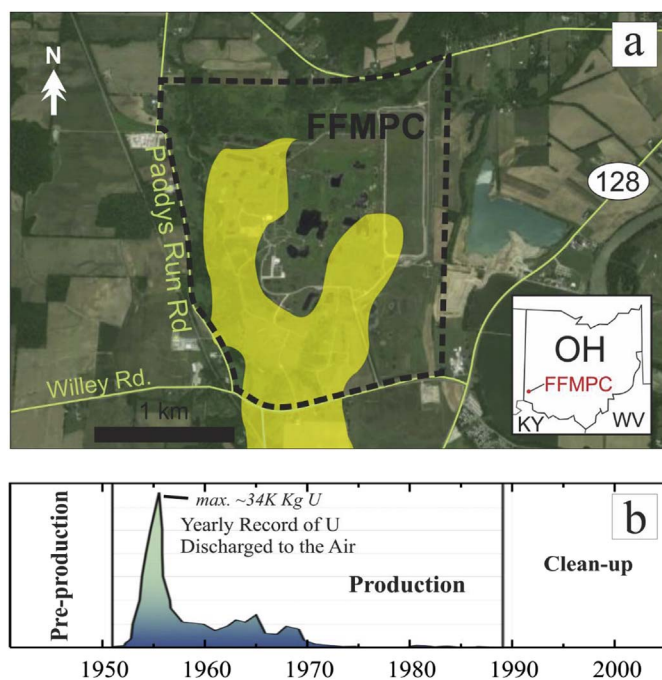
of contamination. These samples preserve high concentrations of U within 5 km of the site, and evidence of anthropogenic U up to 30 km away from the site that is identified as being sourced from airborne particulate matter (Conte et al., 2017, 2016; Widom and Kuentz, 2010b). Additionally, lichens (which act as passive atmospheric particulate filters) collected < 3.5 km from the FFMPC record non-natural U isotope compositions (Tortorello et al., 2013). Tortorello et al. (2013) analyzed a sediment core from an impoundment approximately 7 km from the site. It was found to preserve a record of known U releases revealed by elevated U concentrations in conjunction with non-natural U isotopic compositions. These studies at the FFMPC along with the well-documented history of contaminant release provide the ideal framework for investigating the use of tree rings as records of nuclear material dispersal.

This study tests whether different deciduous tree species act as reliable temporal archives of U contamination. Semi-ring porous (slippery elm, white ash) and diffuse porous (black walnut) species were analyzed by thermal ionization mass spectrometry (TIMS) for U abundance and U isotope ratios. Tree ring U compositions were compared with historical records and a sediment core collected near the FFMPC to assess U mobility within the trees.

## 2. Materials and methods

Tree species were selected based on the availability of trees that predated production at the FFMPC. A 5 mm increment borer was used to

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**Fig. 1.** a Google Earth satellite image of the former FFMP site. FFMP land borders are dashed. Shaded region shows extent of  $> 20 \mu\text{g U/L}$  groundwater plume (1988–1993) as modeled by [Sidle and Lee \(1996\)](#). b Generalized timeline of FFMP history including a record of U released into the air ([FEMP, 1998](#)). Modified after [McHugh et al. \(2015\)](#).

survey over 25 individual trees in the vicinity of the FFMP in order to find specimens that had rings older than 1951. Trees that met this criterion were then sampled for analysis using a battery-operated drill with a 12.5 mm internal diameter incremental borer. Black walnut (UCF13-04) and slippery elm (UCF13-05) cores were collected in September 2013. These trees were sampled  $< 0.5$  km from the FFMP site margin, within approximately 50 m of one another, and above the contaminated groundwater zone ([Fig. 1a](#)). The white ash tree, located approximately 3 km northeast of the FFMP, was cut down in September 2015 and a cross sectional slab was collected from the felled log. Bark from the white ash and slippery elm was stripped by hand from the side of the trunk facing the FFMP in October 2014 and May 2015, respectively. A plug of East-facing black walnut bark was collected from the intact core. All sample materials were frozen for storage, then dried at  $60^\circ\text{C}$  for 24 h before further handling.

The cross-section sample of white ash was trimmed using a table saw to a  $4 \times 4 \times 23.5$  cm block. All surfaces were then smoothed using a hand plane with a stainless steel blade. A smooth, flat surface was created on the two other cores with a carbonized steel chisel blade that eliminated approximately 25% of the exterior of each core. These surfaces were then analyzed at The College of Wooster Tree Ring Lab where rings were counted, and their widths measured with a Velmex Tree Ring Measurement System to a one micron (0.001 mm) resolution and then crossdated with a composite living ring-width series from SW Ohio (SWO). The black walnut core measured 13 cm in length with 70 rings; the slippery elm sample was 11.5 cm with 77 rings; and the white ash tree core was 23.5 cm long with 81 rings. Ring widths were crossdated with the SWO ring-width regional series made up of multiple species of dated, living trees in southwest Ohio. The quality control software program COFECHA ([Holmes, 1983](#)) was used to assess the quality of the dating. A floating ring-width chronology was internally crossdated among four ring-width series (one from each of the slippery elm and black walnut, and two from the white ash cross section) and had a mean intercorrelation of 0.49. This intercorrelation is highly significant with a P-Value  $< 0.00001$  and an overlap of 88 years with the broader SWO dataset. These statistics along with the known

sampling year of the trees generates high confidence that the rings are dated accurately. Based on the year of sampling (i.e., 2013 for the elm and walnut), known marker years, the relative dating of the floating ring-width series, and the crossdating with the regional SWO master, the outer ring date of 2007 was determined for the white ash and that there were no missing or false rings in the black walnut or slippery elm. The outer ring date of the white ash of 2007 is the death date of the tree, which was killed by the emerald ash borer in 2007. Accordingly, the black walnut core dates from 1943 to 2013, the slippery elm from 1936 to 2013, and the white ash from 1926 to 2007. Discoloration of the wood in the center of the white ash tree suggested the onset of decay and, as a result, the innermost rings (1926–1935) were not analyzed.

Details of sample preparation for chemical analysis are given in [McHugh et al. \(2015\)](#) and briefly summarized here. Each dated wood sample was dusted with compressed air to remove visible debris, such as bark or wood particles that may have been deposited during sample extraction and handling. Then, the samples were inspected under a binocular scope to ensure that particles had been removed from pores in the wood's surface. The samples were segmented into 3–5 ring sections using carbonized and stainless steel chisels. Wood and tree bark from each tree was ashed and dissolved in 8N nitric acid.

Dissolved wood and bark were split into a 10% aliquot for U concentrations and a 90% aliquot for isotopic composition. The 10% concentration aliquot was spiked with  $^{233}\text{U}$  for isotope dilution analysis. The U concentration of each sample is presented relative to the ash weight (i.e.  $\mu\text{g U/kg ash}$ ) rather than dry weight of the wood. This approach was selected because of the difficulty handling small amounts of wood ash ( $< 10$  mg), which is prone to dispersal by static charge, and inevitably some loss occurred during transfer between vials. Concentrations relative to wood ash are therefore the most accurate, as we know precisely the mass of ash that was dissolved. Nevertheless, very similar trends are observed for both dry wood and ash concentrations versus time; estimated dry weight concentrations are given in [Supplemental Table 1](#). Sample solutions were loaded on to AG1-X8 anion exchange resin and U was purified in two column steps ([McHugh et al., 2015](#)). Uranium concentrations and isotopic compositions were analyzed at Miami University on a Thermo-Finnigan Triton multi-collector TIMS with a retarding potential quadrupole energy filter. Approximately 0.3–11 ng of U from tree ring samples were loaded onto double rhenium filaments for analysis of isotopic compositions. Reproducibility and accuracy were monitored by analysis of NBS U005a and U145b. NBS U005A ( $n = 48$ ) was measured at  $^{235}\text{U}/^{238}\text{U} = 5.13(4) \times 10^{-3}$  (certified value =  $5.09 \times 10^{-3}$ ),  $^{234}\text{U}/^{235}\text{U} = 3.49(21) \times 10^{-5}$  (certified value  $3.41 \times 10^{-5}$ ), and  $^{236}\text{U}/^{238}\text{U} = 1.25(26) \times 10^{-5}$  (certified value =  $1.19 \times 10^{-5}$ ). Average values for U145b (derived from CRM 112-A) ( $n = 31$ ) are  $^{235}\text{U}/^{238}\text{U} = 7.30(4) \times 10^{-3}$  (certified value =  $7.25 \times 10^{-3}$ ) and  $^{234}\text{U}/^{238}\text{U} = 5.37(15) \times 10^{-5}$  (certified value  $5.28 \times 10^{-5}$ ). A correction factor of 0.994 and 0.985 was applied to measured  $^{235}\text{U}/^{238}\text{U}$  and  $^{234}\text{U}/^{238}\text{U}$ , respectively, based on U145b analyses. Errors (2-sigma) on individual analyses are estimated based on the external reproducibility of U005A unless the individual run error exceeded that of U005A, in which case the within-run error is presented. No replicates of wood were analyzed because of limited sample size and low U abundances. Total procedural blanks were  $< 60$  pg U.

### 3. Theory

#### 3.1. U isotope systematics

Anthropogenic processes modify the relative abundance of naturally occurring U isotopes and, as a result, their relative abundances can be used to trace nuclear contamination in the environment. Uranium-235 and U-238, the major isotopes of U, are the most abundant and account for 99.995% of total environmental U. Uranium-234 comprises the

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