RUTHINIUM CONTAMINATION ON POLLEN ACCUMULATION RATES:

A PRELIMINARY CASE STUDY FROM WILLIAMS LAKE, WA.

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ABSTRACT

The impacts of radionuclides in the environment and on human health have been studied in detail following the 1986 nuclear meltdown of Chernobyl and the more recent 2011 fallout from Fukushima. Research into biological impacts of radionuclides exposure increased following the 2017 undeclared radio ruthenium (^{106}Ru) release in Eurasia (Masson et al., 2020). However, the ecological impacts of ruthenium on pollen abundance remain to be studied. In this paper, we analyze the environmental impacts of the 1954 ruthenium release event that contaminated vegetation in Eastern Washington ("Hanford Officials," n.d.). A 50-cm frozen sediment core was collected from the deepest portion of Williams Lake, WA (Power, M., 2019). A 15-cm subsection of the core, beginning at the mud-water interface, was sampled at millimeter increments and provides detailed information on pollen production during the 20th century. Pollen samples were used in conjunction with aerial photography and satellite imagery to reconstruct vegetation assemblages from 1920-2019 AD. A volcanic tephra layer, located at 16-cm depth, was linked to the 1980s' Mt Saint Helens eruption (Anderson et al., 1984). Changes in 20th century pollen production were compared to climatic and anthropogenic influences to explore potential impacts on vegetation of radionuclides, including ^{103}Ru and ^{106}Ru. This study identifies both the potential short-term (years) and long-term (decades) impacts of radionuclide fallout on pollen abundance.

INTRODUCTION

The Hanford Nuclear site encompasses 1,517 square kilometers along the Columbia River in Southeastern Washington (cite). The site was in operation throughout WWII and at the beginning of the Cold War as a major producer of plutonium for atomic weaponry and housed the world's first full-scale nuclear reactors and chemical reprocessing plants (Gephart, 2012). Nuclear waste, both intentionally and accidentally, was released from the Hanford Nuclear site on multiple occasions, impacting communities that lived downwind and downstream from these releases (Gephart, 2012; Stenehjem, 1989). In 1986, due to the Freedom of Information Act, the Hanford Nuclear Site released thousands of previously classified documents regarding the operational history and storage of toxic waste at its facilities (Stenehjem, 1989). From these reports it was revealed that millions of curies of radiation and billions of gallons of hazardous liquids had been released into the environment across the Columbia Basin. Of the release events and spills revealed by the Hanford documents, the ruthenium release in January of 1954 was one of the largest ("Hanford Officials," n.d.). The release contaminated 10,500 km^2 of

Eastern Washington. Concentrations of ruthenium (^{103}Ru and ^{106}Ru) recorded near the source were >10-1 µc/gm of vegetation, with concentrations of ruthenium recorded 10-3-10-4 µc/gm over a hundred miles away from the initial site ("Figure C-20," n.d.). Williams Lake is located 160km (100 miles) northeast from the Hanford site and near the extent of the ruthenium plume event (see fig 1).
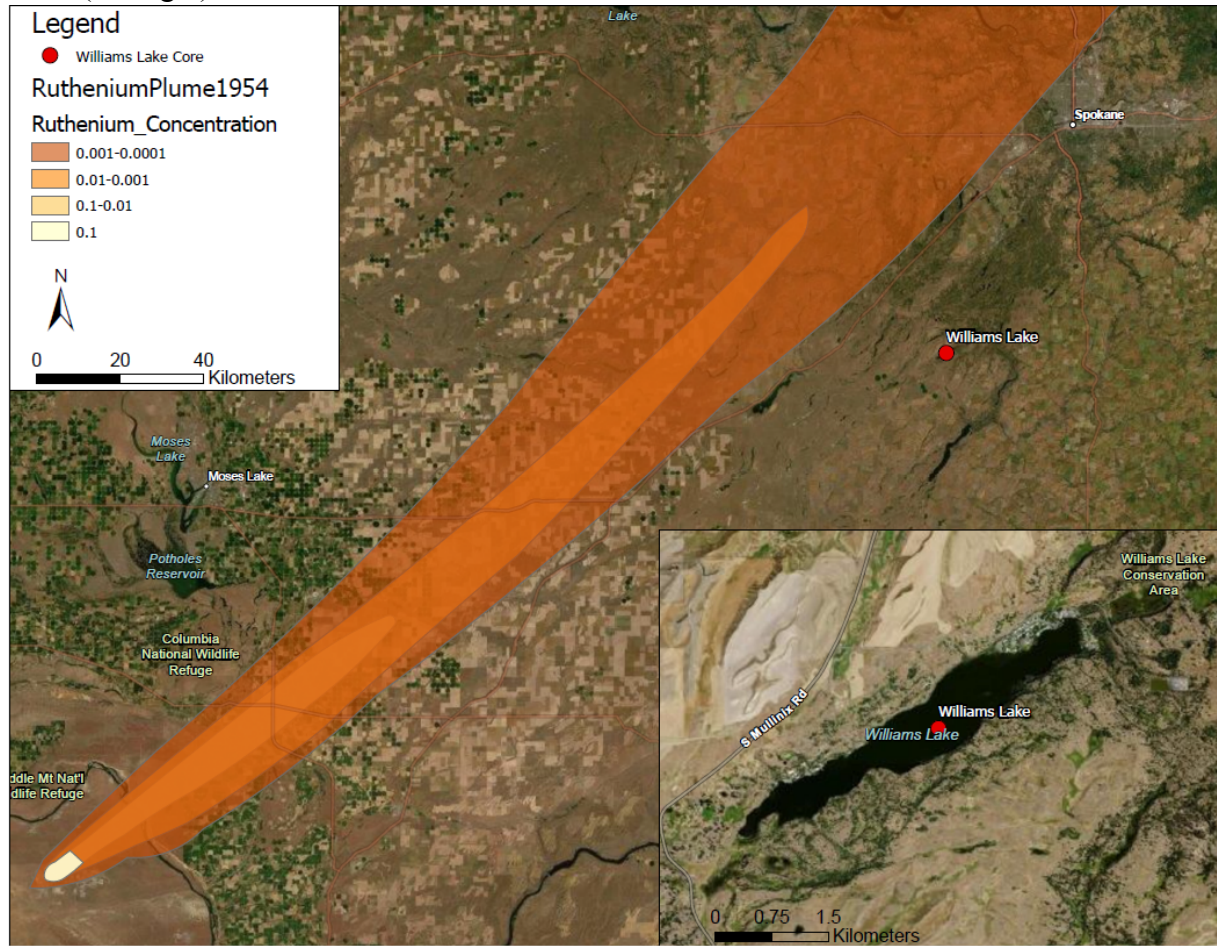
Fig 1. Estimated extend of 1954 Ruthenium (ru103-ru106) plume cloud in relation to Williams Lake coring site (47.326409°, -117.684750°). (Power, 2019; Pecchia-Bekkum, 2020).

Vegetation is cited as the primary source of ruthenium contamination found in biological systems through direct contamination of plants and subsequent transport through trophic levels (Ridisk et al, 1955; Pröhl, 2009). This can occur through direct airborne contamination on vegetation as well as absorption through roots in contaminated soils (Brown, K. W., 1976, p. 11; Pröhl, 2009). In their study, Iwashima & Yamagata (1966) note that radio ruthenium (Ru103, Ru106) is not easily retained by soils. As such, contamination of water systems via runoff is higher for radio ruthenium than cesium, a less mobile radionuclide. More recent studies indicate that due to the restricted mobility of ruthenium in soils, contamination of trophic levels through this process are rare. Of the ruthenium that is absorbed by soils, as much as 99% of ruthenium is held in the roots of the plants and not easily transported through aboveground trophic levels (Zuba et al. 2020).

SITE DISCRIPTION

Williams Lake is an endorheic lake within the Cheney-Palouse tract of the Channeled Scablands in Spokane County, Washington (Nickmann, 1979). The dominant vegetation surrounding Williams Lake is ponderosa pine (*Pinus ponderosa),* shrubs (Asteraceae *Artemisia),* and grasses (Poaceae*).* Sedges (*Carex rostrata*)*,* bullrush (*Scirpus acutus*)*,* willow (Salix), quaking aspen (*Populous tremuloides),* and dogwood (*Cornus stolonifera*) were noted in previous work by Nickmann (1979). The region lies in the transition zone between dry, sagebrush-grasslands in the Columbia basin and forested Selkirk and Bitterroot mountain ranges to the East ('Ponderosa Pine Woodland", 2013). The Columbia Basin is a semi-arid cold desert with annual precipitation as little as 3-6in on the leeward side of the Cascades (Washington Native Plant Society, 2019). A combined modified maritime and continental climate bring milder winters due to the influence of Pacific air masses (Nickmann, 1979). While precipitation occurs year-round, the primary influx of moisture occurs between September and April, with dry conditions prominent during the summer months (Nickmann, 1979; Washington Native Plant Society, 2019). Fire seasons tend to peak in the late summer and early fall in these modified shrub-steppe ecosystems (Washington Native Plant Society, 2019).

METHODS

On September 24, 2019, a 50cm core was collected from Williams Lake using a freeze core recovery system (Power, 2019). The deepest portion of the lake (47.326409°, -117.684750°) was targeted for coring. During initial preparation of the core, a single radiocarbon date was collected to construct the preliminary age-depth model. The bulk sample was collected at a depth of 49.5-50.5 and given an initial age of 610 BP (see Table 1).

Table 1. Radiocarbon Date for Williams Lake, WA.

UGAMS#	Sample ID	Material	δ13C, ‰	14C age, years BP	+/-	pMC	+-
45108	WLFC 19-49.5-50.5	Bulk sediment	-27.81	610	20	92.7	0.23

Samples were cut at 2mm intervals along the length of a 15cm subsection of the Williams Lake core (see fig 2). Seventeen samples were brought to the Garret Biology Wet Lab at the Natural History Museum of Utah and processed using standard acetolysis preparation. Emphasis was paid to sample depths 28-30mm and 48-50mm, having estimated age-depth of 1981 and 1954, respectively. Samples surrounding these dates were selected in tandem with modern pollen samples to construct changes in vegetation for the past 100 years. All surfaces were cleaned using distilled water and a sterile tissue between samples to limit possible cross contamination.
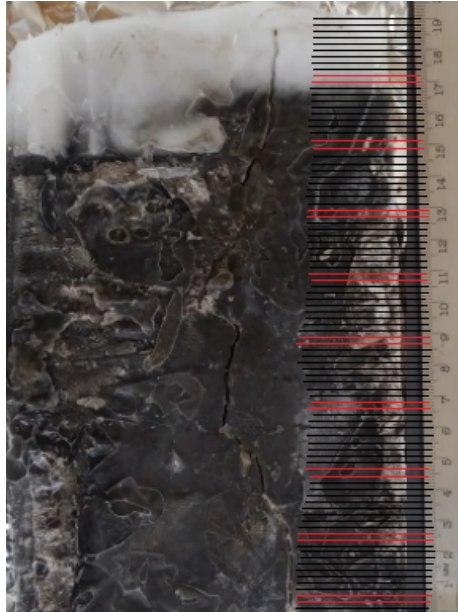
Figure 2. Top 15cm of Williams Lake, WA. Core. Markings indicated 2mm intervals. Red lines denote 10mm intervals.

RESULTS & DISCUSSION

Pollen assemblages were analyzed for eleven samples from the Williams Lake sediment archive. AP/NAP (arboreal/non arboreal) and absolute pollen abundance (PAR) were reported for each sample. Notable peaks in Pinaceae are observed at sample depths 58-60mm (~1941AD), 22-24 mm (~1984AD), and 2-4 mm (~2016AD). Sample depths 50-26 mm indicate little change in Pinaceae abundance but a distinct decrease from surrounding samples, ~35% compared to that of ~49% of total pollen for each sample.

Increase in Poaceae and Amaranthaceae pollen abundance during the early 1980s, ~1981-1984 (26-30mm), is potentially a response to the 1980s Mt Saint Helens Eruption. A notable decrease in Asteraceae during the same period points to high disturbance, as weed types and grasslands replace native vegetation. Due to its short lifecycle, many grass species benefit from the short-term increase of soil nutrients following volcanic eruptions (Urrutia, et al. 2007; Engels, S. et al., 2015). This may explain it subsequent decrease at depths 22-24 mm following a recovery of Pinaceae. Betulaceae remains stable throughout the pollen record, though a notable fluctuation of ~1.5% occurs throughout the record. Cyperaceae percentages are low throughout the pollen record. However, notable increases of Cyperaceae pollen at depths 28-50 mm (~1984-1954) may be an indication of drier conditions as lake levels recede and marshland expands around the lake margin (Anderson, M., 2008). Cupressaceae ranged in our pollen record from 3-8%, with notable increases at depths 2-4 mm, 26-28 mm, and 74-76 mm (~2016, ~1981, & ~1920) (see Fig 3).
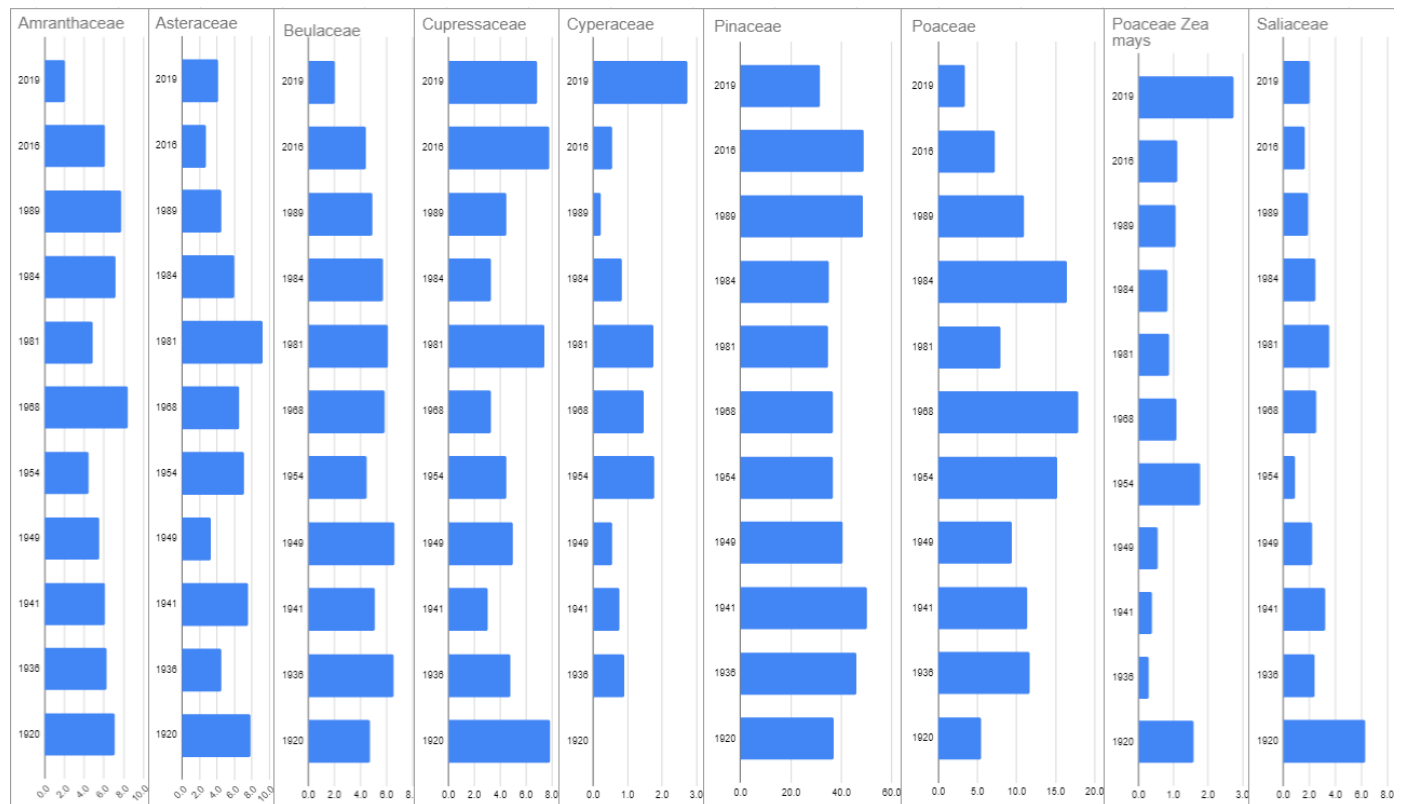
Figure 3. Pollen percentages for selected pollen groups. Samples corresponding to Mount St. Helens Eruption (1980) and Ruthenium plume leak (1954) are highlighted in blue and red, respectively. Fabaceae, Brassicaceae, and Apiaceae omitted due to high uncertainty in pollen identification. Age gaps between samples range from 3-27 years. Absolute age-depths are represented by Y-axis and may indicate rapid changes due to gaps in chronology. X axis represents pollen percentages by pollen type. Note* a unique x-axis for each pollen category is present.

Pollen fluctuation was compared to moisture and temperature graphs from Spokane County, WA. Seedling germination of *P. ponderosa* is dependent on open canopies, mineral soils, and sufficient moisture within the first two growing years. May-June precipitation is a significant indicator of seedling success and establishment (Fryer, 2018; McDonald & Fiddler, 1989.). Late spring moisture and dry summers have been associated with high cone production (Fryer, 2018). From this, we assume that high cone production years are synchronized with high pollen production for *P. ponderosa* and is an indication of heavy spring moisture and summer drought. From this assumption, we would expect to see high *P. ponderosa* pollen production in 2017, 1974, 1953, 1940, and 1932 (see fig 4). This assumption corresponds well to our PAR analysis, indicating high *Pinus* pollen production in ~1940 and ~1981. Rapid increase in *Pinus* pollen production occurred between ~1936 and ~1941. However, ~2017 and ~1954 both show marked decreases in *Pinus* pollen production contrary to our assumption of a high pollen yield. Looking at the relative pollen percentages (fig 3), there is a notable increase *Pinus* pollen (~50%) In ~1941 compared to 1954 (38%), which may account for some of this variability. This does not explain the dip in ~2016, though this may be an indication of other climatic variables such as temperature. Precipitation for ~1954 was in-line with the 1970-2000 climate average. Assuming that this constitutes an average pollen production year, we can rule out climate as a significant contributing factor to pollen variance observed during this time period.
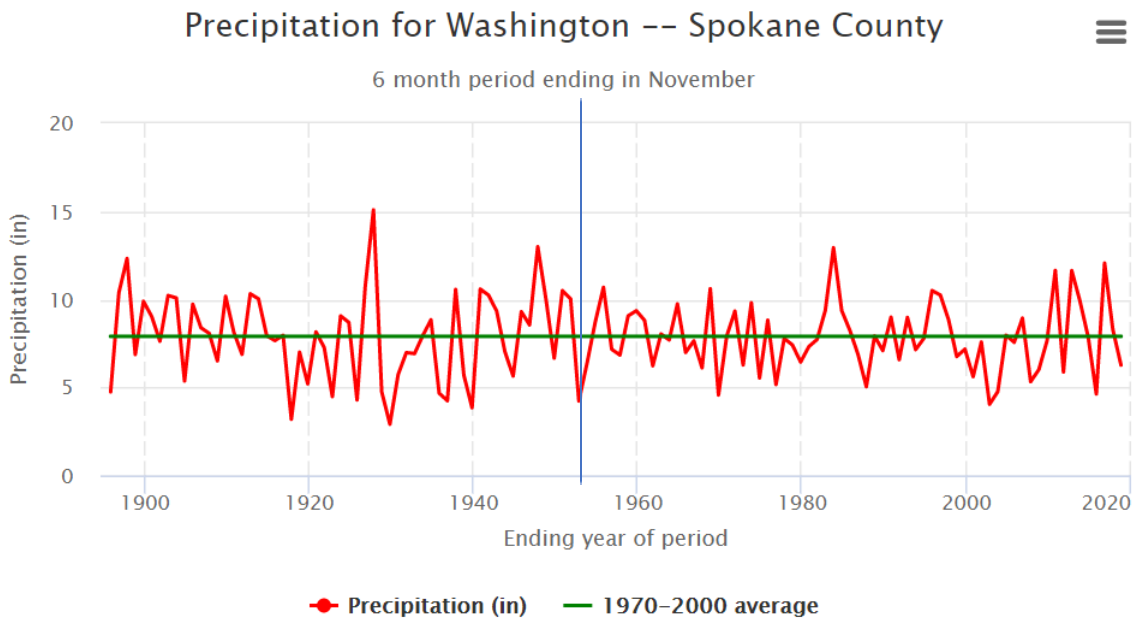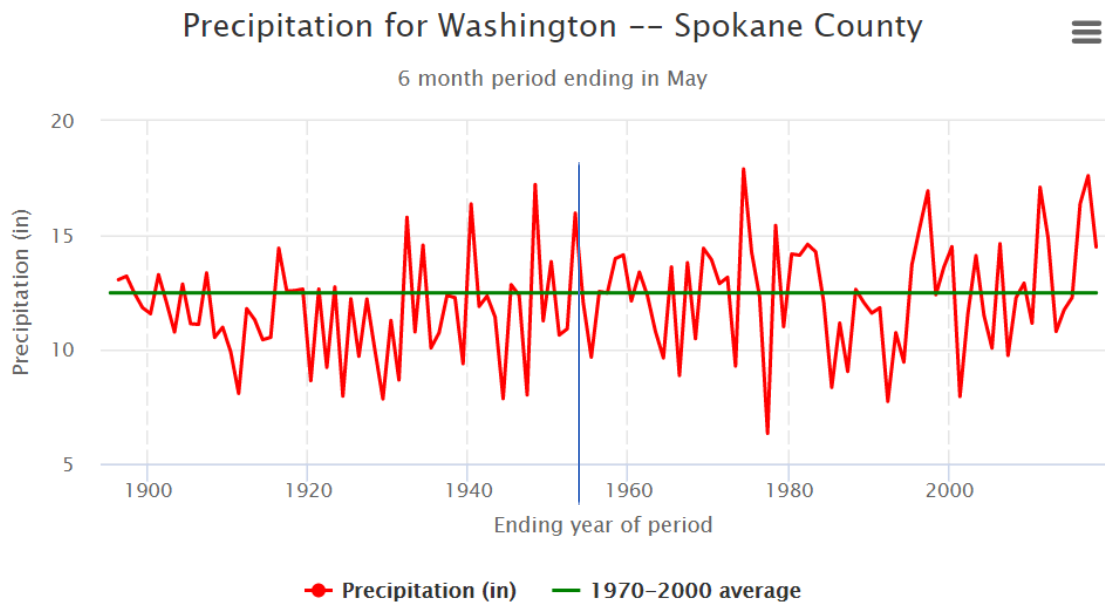
Fig 4. Precipitation averages for Spokane County, WA with 1970-2000 running mean. Top: aggregated precipitation data for winter and spring precipitation (December – May). Bottom: summer and fall precipitation (June-November). Blue line indicates 1954 AD.

FUTURE WORK

Anthropogenic land-use change and fire history, not covered in detail in this report, would provide greater context for understanding natural pollen variance. Work has gone into exploring the human impacts of land use from archaeological and historic records. Charcoal counts would provide greater detail into the fire history for the region and provide a means to improve our age-depth model. A greater number of samples, would enable us to reconstruct vegetation response in greater detail as well as provide a better understanding of how vegetation may respond to anthropogenic activity and biological pollutants.

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Figure C-20 Ruthenium Contamination on Vegetation of Eastern Washington, January, 1954. (n.d.) IInternal PNNL Report.

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