Journal of Environmental Radioactivity 99 (2008) 1355–1359

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Short communication

Radioactivity in fossils at the Hagerman Fossil Beds National Monument

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ARTICLE INFO

Article history:
Received 14 November 2007
Received in revised form 11 February 2008
Accepted 12 February 2008
Available online 28 April 2008

Keywords:
Fossils
Hagerman Fossil Beds
Uranium
Natural radioactivity

ABSTRACT

Since 1996, higher than background levels of naturally occurring radioactivity have been documented in both fossil and mineral deposits at Hagerman Fossil Beds National Monument in south-central Idaho. Radioactive fossil sites occur primarily within an elevation zone of 900–1000 m above sea level and are most commonly found associated with ancient river channels filled with sand. Fossils found in clay rich deposits do not exhibit discernable levels of radioactivity. Out of 300 randomly selected fossils, approximately three-fourths exhibit detectable levels of natural radioactivity ranging from 1 to 2 orders of magnitude above ambient background levels when surveyed with a portable hand held Geiger-Muller survey instrument. Mineral deposits in geologic strata also show above ambient background levels of radioactivity. Radiochemical lab analysis has documented the presence of numerous natural radioactive isotopes. It is postulated that ancient groundwater transported radioactive elements through sand bodies containing fossils which precipitated out of solution during the fossilization process. The elevated levels of natural radioactivity in fossils may require special precautions to ensure that exposures to personnel from stored or displayed items are kept as low as reasonably achievable (ALARA).

1. Background

The official Idaho State Fossil, the 'Hagerman Horse', was discovered at Hagerman Fossil Beds National Monument (HAFO) in the late 1930s and it is radioactive. Radioactive fossils at the Monument were noted in the National Park Service (NPS) Conserve O Gram newsletter in 1993 describing the importance of required monitoring for radiation (NPS, 1993). The 4394 acre Hagerman Fossil Beds National Monument is located 90 m southeast of Boise near the Snake River town of Hagerman in south-central Idaho. The fossil beds were commissioned in 1988 to preserve, protect, and provide for study the Pliocene fossil assemblage. The site has been described in several publications (Malde and Powers, 1958; McDonald, 1993; McDonald et al., 1996) and contains numerous fossil sites including the well known Hagerman Horse Quarry where more than 200 extinct zebra-like horse fossils, *Equus simplicidens*, have been recovered from just one location (Carpenter, personal communication). More than 1000 individual fossil sites have been mapped with mostly terrestrial mammals represented from the floodplain environment of ancient Lake Idaho that deposited the Glenns Ferry Formation. At least 111 radioactive fossil sites have been documented at HAFO from this study, including the 'Hagerman Horse Quarry' (Farmer and Christensen, 2007). Although radioactive accumulation in fossils is well known, and indeed has been reported at other locations (Smith and Bradley, 1954; Madson, 1983), it was not until 1996 when elevated levels of radioactivity were documented in fossil specimens at HAFO by park service staff (Farmer and Knowles, 1996). This prompted further investigation into the nature and hazards from these specimens.

2. Geologic association

A preliminary scoping study to determine presence, absence and spatial distribution of radioactivity revealed that most of the radioactive fossils are found in ancient river channels filled with porous fine to coarse grained sand. Generally, the presence and level of radioactivity dissipate in the upper and lower stratigraphic elevations of stratigraphy of the plateau by about 30 m (100 ft). The presence and levels of radioactivity increase in fossils deposited in sand units and decrease in fossils found in clay rich deposits. The floodplain deposited clay units typically exhibit drab colors of olive green capped by a dark brown finely laminated clay. These characteristics infer a reducing environment associated with the clay units during deposition. Conversely, the river sand channel that the 'Hagerman Horse Quarry' is deposited exhibits oxidizing conditions inferred from a red oxide colored portion of the sand unit but not all sand units within the Monument exhibit this red oxide coloration.

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doi:10.1016/j.jenvrad.2008.02.004

The presence of radioactivity appears to dissipate in the upper elevations (Tuana Gravel Fm.) based on testing a camel fossil, pine species log, and a $CaCO_3$ layer which exhibit near background levels of radioactivity. It also decreases in the lower elevations of the stratigraphy based on testing the base of an unnamed basalt at an elevation of 869 m and minor numbers of fossils. The Shoestring basalt (Malde and Powers, 1962) base, located at an elevation of 975 m, exhibits post-eruption secondary mineralization with FeO_2. Uranium was probably incorporated into this mineralization process from groundwater under the same conditions as the fossils. Porosity appears to play a role in the hypothetical transport of dissolved uranium flowing within ancient groundwater systems through porous geologic deposits. Therefore, depositional mechanisms are a significant factor in the spatial distribution and level of radioactivity found in the fossils.

The original source for the uranium is unknown but it is likely incorporated into the fossil through a process of chelation. When the fossil is transitioning from organic compounds into minerals, the organics attract radioactive elements. The uranium fills a location in the mineral lattice structure normally held by some other common ion, such as calcium. Fossil dentitions appear to have higher concentrations than other parts of the fossil. Possible sources for the uranium may include silicic volcanic ash layers, hydrothermal systems or sediment parent rock such as the Idaho Batholith granites, possibly near the town of Sun Valley.

3. Survey and mapping data

Elevations of fossil sites were derived from two different methods. Older surveys used a 'Criterion' laser transit tied into benchmarks placed by the US Bureau of Land Management Cadastral division in 1993 to calculate coordinates to an elevation accuracy of 0.3 m. The second method used a combination of horizontal coordinates from a Trimble Pro XR with corrected data to produce horizontal accuracy of about 1 m. These points were plotted in ArcGIS 8 with five-foot contour line intervals that have a vertical accuracy of 0.10 m. Fossil site elevations were interpolated based on these contour lines and GPS derived points thereby eliminating the use of GPS elevations which are typically an order of magnitude greater error than horizontal coordinates. Then all of the sites were grouped into 10 m elevation zone categories shown in Fig. 1 as the horizontal bar graphs.

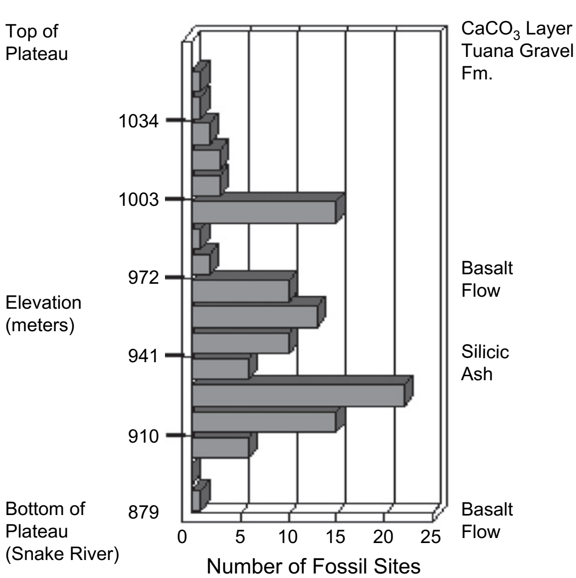

Fig. 1. Vertical frequency graph showing 111 radioactive fossil sites (*x*-axis) at HAFO relative to elevations (*y*-axis in meters) with three peaks at 925, 960 and 990 m.

A preliminary map (Fig. 2) and vertical frequency distribution graph (Fig. 1) were developed using GIS to aid this scoping investigation with identifying the vertical and horizontal distribution of radioactivity. The elevation of the Snake River is 854 m and the top of the plateau is about 1052 m. The circles denote radioactive fossil sites with different levels of radioactivity in counts per minute (cpm) as determined with a hand held portable Geiger-Muller survey meter and described in more detail in Section 4. The lack of sites in the northern and southern 25% of the Monument is due to less fossil exposure from erosion, more vegetation and ancient landslides. The sinuous dashed line on the left side of Fig. 2 is a $CaCO_3$ layer that exhibits only ambient background levels of radioactivity. The $CaCO_3$ layer is located at the top of the formation at an elevation of 1036 m. The black lines in the south area represent the Peter's Gulch silicic volcanic ash which measured 70–120 cpm on a portable Geiger-Muller survey meter, several fold greater than measured backgound levels of 20–50 cpm. In the north area is the Shoestring basalt flow, positioned at an elevation of 975 m, mid-level within the stratigraphic section and noted with a black hachured line. The rubble iron oxide base (Fig. 3) measured 400 cpm while the massive part of the basalt above the rubble base is near ambient background levels of 20–50 cpm on the same instrument. The base of the lower unnamed basalt at an elevation of 869 m (2850 ft) also measured at ambient background levels and is located at a low elevation in the formation. The vertical distribution (Fig. 1) indicates that most of the radioactive sites are located between about 900 and 1000 m elevation. It is unknown why radioactive depositions appear to be concentrated between these elevations, but they could be related to prehistoric rainfall, water flow patterns or proximity to silicic volcanic ash layers. There are three peaks at 925, 960 and 990 m elevation and the cpm values generally ramp up and down from the two lower elevation peaks while the upper peak at 990 m is more analogous to a spike. It is presumed that the spatial distribution of the known radioactive fossil sites will correlate to the pattern of all sites.

4. Radioactivity measurements

Initially, elevated levels of radiation were detected in fossil specimens at HAFO using a portable Geiger-Muller (GM) survey instrument equipped with a thin window pancake detector with a window thickness of 1.5 mg cm^{-2} (Fig. 4).[1] A preliminary evaluation was performed at the University of Idaho which suggested uranium in concentrations of up to several weight percent. Recently, it was determined that mining claims were filed during 1950s within the Monument area for geologic deposits containing radioactive fossils.

Radon-222, a product of the uranium-238 $(4n+2)$ decay chain, has been measured inside fossil storage cabinets with a maximum of 544 pCi L^{-1} $(2.0 \times 10^4$ Bq $m^{-3})$ and an average of 436 pCi L^{-1} $(1.6 \times 10^4$ Bq $m^{-3})$. Ambient radon concentrations in the fossil storage room where staff work have been recorded by National Park Service staff (Farmer and Christensen, 2007) with a mean of 12 pCi L^{-1} and a maximum of 26 pCi L^{-1} (960 Bq m^{-3}), which corresponds to approximately 2.7 working level (WL). Thus the potential exists for significant radiation exposure to personnel working in this location for an appreciable portion of the workday, or to members of the general public, and in particular children, who may be casual visitors to the site, indicating a need for safety precautions to ensure that exposures are kept below permissible levels and consistent with the ALARA (as low as reasonably achievable) philosophy. The data collected were under typical working conditions

[1] Technical Associates Model TBM-3S.

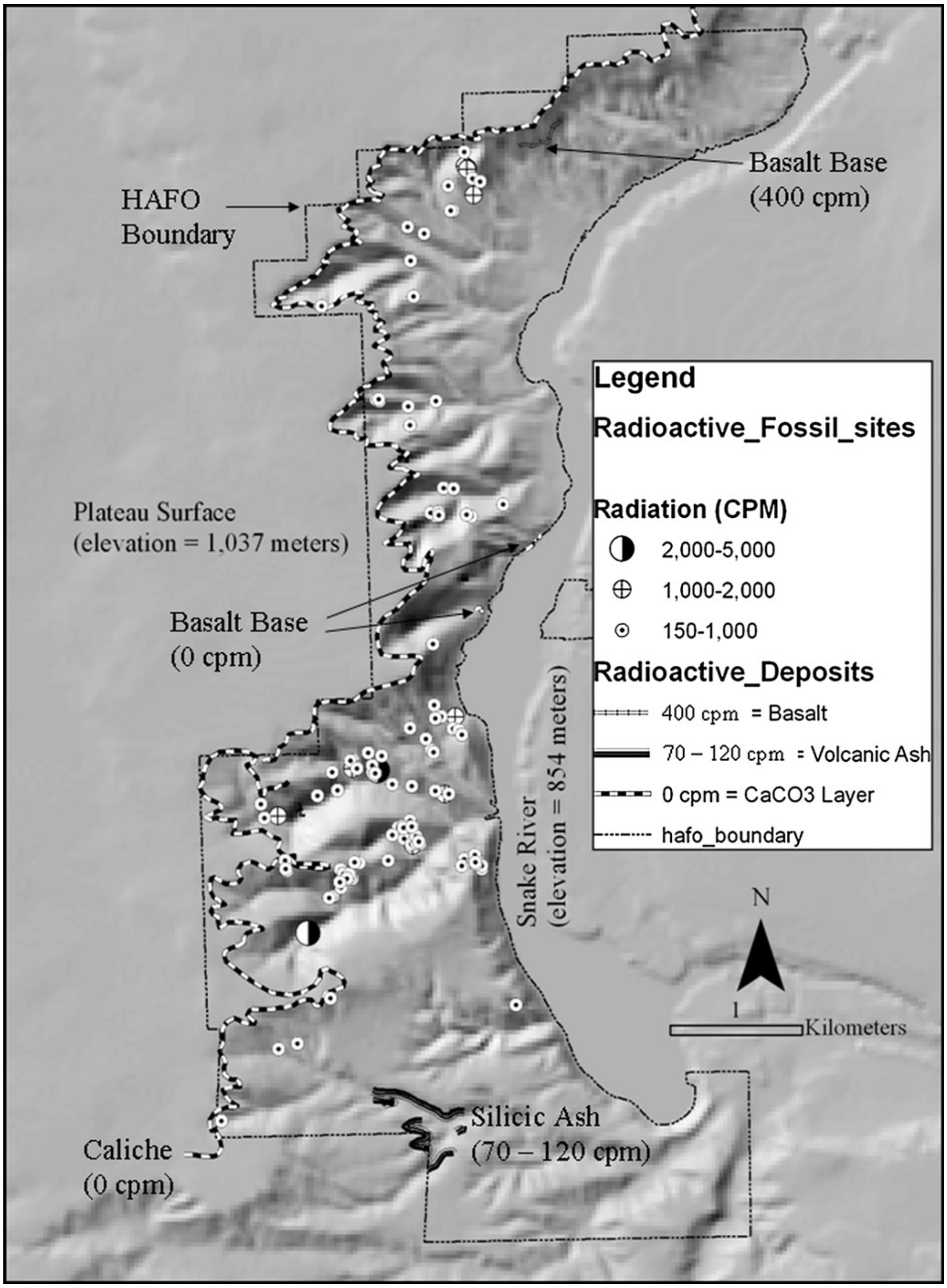

Fig. 2. General location map showing horizontal distribution patterns of radioactive fossil sites. Radioactivity levels are relative and not have been corrected for background, which ranged from near 0 to 25 cpm.

with no effective outside ventilation and the environmental control system functions to heat or cool the building according to thermostat setting. It is recommended that future studies measure the length of time of exposure to humans in addition to concentrations of radon and the decay products of radon. It was common, prior to year 2006, for staff to work directly in the storage room with fossil cabinet doors open and little or no outside ventilation occurring.

During 2005, more than 300 fossil specimens were tested with a portable Geiger-Muller survey meter in contact with the sample (Fig. 4). Radiation levels greater than twice the ambient background levels of 20–50 cpm were detected in three-fourths of the samples tested; 136 (45%) exhibited levels greater than 500 cpm with one specimen (cat. #256) measuring 7000 cpm.

A preliminary radiochemical analysis on a single fossil specimen revealed the presence of a number of naturally occurring radionuclides from the uranium ($4n+2$) series nuclides (U-238, Ra-226, Pb-214, Bi-214, and Pb-210) and the neptunium ($4n+3$) series (U-235, Ra-223, and Pa-231). A follow-up study was made on the radioactivity content of 10 different fossilized bones taken from various locations at the Hagerman site. The radioactivity content of the samples was determined by external gamma ray spectroscopy at the In Vivo Radioassay and Research Facility in Richland, WA, using a 38 cm^2 high purity germanium detector in a lead shielded copper

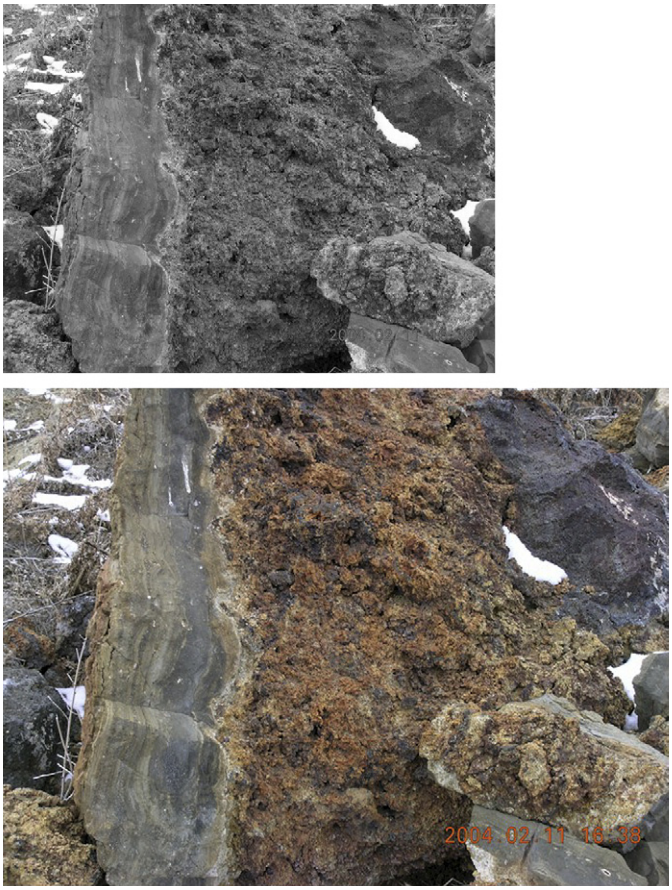

Fig. 3. Iron oxide rubble base of the mid-stratigraphic section basalt flow (elevation 3200 ft) measured at 400 cpm (including ambient background radiation).

lined room. The samples were counted for 10 or 20 min at a centroid to detector distance of 35 cm, sufficiently great to assume point source geometry. Calibration was accomplished with standard U-238 and Ra-226 sources. The Ra-226 content of the samples was determined from the 186 keV photon associated with Ra-226 decay. Uranium content was obtained by assuming secular equilibrium U-238 and Th-234 and determining the activity from Th-234 from the mean of the values calculated using 63 and 93 keV lines associated with the decay of Th-234. The results are shown in Table 1.

In eight of the 10 samples analyzed, Th-234 was measured at concentrations ranging from 5 to 31 Bq g^{-1} with a mean of 12.1 $\pm$ 7.7. Assuming secular equilibrium between U-238 and Th-234, and the specific activity of 12.8 kBq g^{-1} for natural uranium, this corresponds to an average uranium content of about 1000 ppm. Six of the 10 samples examined exhibited detectable levels of both Th-234 and Ra-226. In this group of six samples, the Ra-226:Th-234 ratio exceeded unity, ranging from 1.09 to 1.41 with a mean of 1.25 $\pm$ 0.11, suggestive of disequilibrium among the members of the uranium decay series beyond Th-234, likely attributable to the greater solubility and thus potential greater leaching of radium although other mechanisms could also be responsible (Faure and Mensing, 2004). In one of this group of seven samples, the Ra:Th ratio was only 0.523, an apparent anomaly with no readily obvious explanation. No detectable activity was observed in the smallest two samples, likely because the activity present was below the detection limits of the system used and the 20 min counting time.

5. Radiological implications

Radioactive fossils have a number of practical implications, both positive and negative. On the positive side, the concentration of natural radioactivity in fossils can serve as an aid to finding and specifically locating and mapping fossils in the field, given suitably sensitive instrumentation, properly used. It can also be used to trace sediments back to parent rock source areas thus delineating paleo drainage patterns. However, on the negative side, radioactivity accumulated in fossils can pose the possibility of a radiation hazard, and care needs to be taken to ensure that exposures from this source are adequately controlled. The NPS recommends that when radon concentrations are above 4 piC L^{-1}, an action program for monitoring must be implemented and that large collections of radon-bearing specimens should be housed in a dedicated space (NPS, 1993). Currently, continuous ongoing radon monitoring has not been implemented by the park paleontology program (Carpenter and Christensen, 2007). Because of the potential for external radiation fields associated with these fossils as well as the emanation and buildup of Rn-222 and progeny, precautions should be taken in handling and displaying these and other fossil samples containing elevated levels of natural radioactivity to maintain exposures as low as reasonably achievable (ALARA). One precaution taken by the NPS was to install a dust evacuation enclosure where fossils are prepared out of the sand matrix.

Collections of radioactive fossils held in storage likely present the greatest potential for exposure, for even a single or few fossils that have acquired a large concentration of natural activity could conceivably produce external dose rates that could result in exposures in excess of permissible occupational limits. Prior to year 2001, the fossil storage was at a different location with different

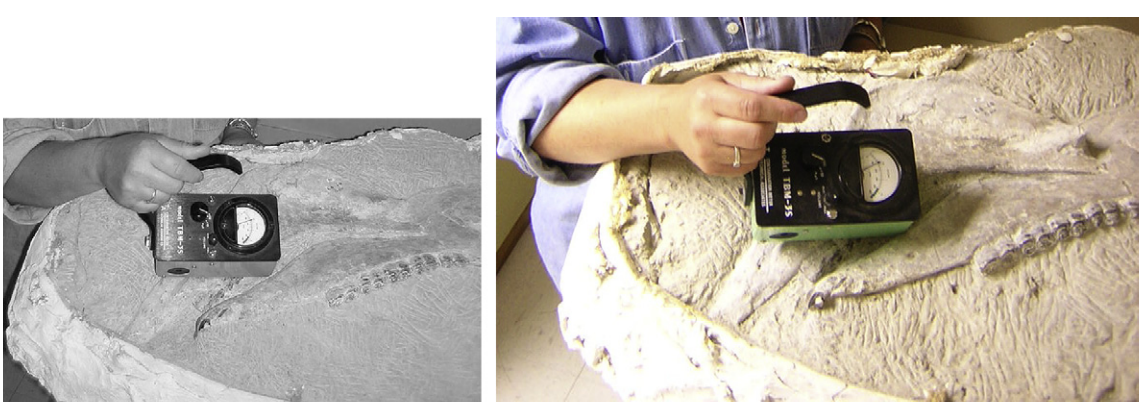

Fig. 4. ‘Hagerman Horse’ mandible fossil monitored with a Geiger-Muller survey meter.

Table 1
Uranium ($4n+2$) series activity in fossilized horse bone samples

HAFO identification number	Weight (g)	Th-234		Ra-226		Ratio
		Activity (Bq)	Concentration (Bq g^{-1})	Activity (Bq)	Concentration (Bq g^{-1})	Ra:Th concentration
251	609	6967 ± 521	11.4	7590 ± 594	12.5	1.09
253						
254						
388	29	224 ± 69	7.7	ND	–	–
391	165	840 ± 101	5.1	440 ± 158	2.7	0.52
2054	471	2361 ± 213	5.0	3337 ± 270	7.1	1.42
2173	102	1434 ± 158	14.1	1947 ± 214	19.1	1.35
2202	98	3007 ± 132	30.7	3740 ± 289	38.2	1.24
2208	301	2728 ± 218	9.1	3286 ± 270	10.9	1.20
2229	392	5237 ± 295	13.4	6087 ± 389	15.5	1.16
2329	25	ND	–	ND	–	–
95–58	24	ND	–	ND	–	–

building structure and significantly fewer quantities of fossils due to a large excavation of 'Hagerman Horse Quarry' fossils 2001 (NPS, 2004) with a recommendation to implement continuous real time radon monitoring. Future data collection should include external dose rate measurements and estimated DAC-hr exposure to provide a solid basis for evaluation of radiological exposures and associated risks. As of May 2006, there have been no dose rate measurements collected under the current fossil storage conditions (Farmer, 2006).

Similarly, displays of radioactive fossils could result in exposures of concern to members of the public, particularly children handling fossils at the 'Hagerman Horse Quarry' (Farmer and Christensen, 2007). Uranium bearing fossils may contain significant quantities of Ra-226, the precursor of Rn-222, and buildup of hazardous concentrations of Rn-222 and daughters in storerooms or other enclosed areas containing radioactive fossils is likely.

6. Summary and conclusions

Naturally occurring radioactivity at elevated concentrations is well documented at Hagerman Fossil Beds National Monument and in fossils obtained from that site. Spatial distribution of radioactive fossil sites occurs primarily between an elevation zone of 900 and 1000 m. They are most commonly found associated with ancient river channels filled with sand, and fossils found in clay rich deposits do not exhibit discernable levels of radioactivity. Approximately three-fourths of a group of 300 fossils from the Hagerman site showed elevated concentrations of natural radioactivity when monitored with a portable Geiger-Muller survey meter. Uranium concentrations in 10 fossil samples quantitatively examined for uranium were typically on the order of about 1000 ppm by weight and a few tens of Bq g^{-1}; radium activity concentrations were typically slightly greater than those of U-238, indicative of disequilibrium between U-238 and Ra-226. Because of the potential for external radiation fields associated with these fossils as well as the emanation and buildup of Rn-222 and progeny, precautions should be taken in handling and displaying these and other fossil samples containing elevated levels of natural radioactivity to maintain exposures as low as reasonably achievable. Specific dose rate information needs to be collected on a long-term consistent basis to fully assess the exposures. Continuous real time radon monitoring is recommended as an integral part of the museum collection management program as recommended by NPS internal reports.

Acknowledgements

The authors are grateful to Bob Willhite, former Park Ranger at Hagerman Fossil Beds National Monument, for his support of this project from 1996 to 2001, Timothy Lynch, Pacific Northwest National Laboratory, for gamma spectroscopy of the fossil samples, and Mary Carpenter for manuscript review and information.

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