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Detection of Bullet Residue in Bone Using Proton-Induced X-Ray Emission (PIXE) Analysis

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ABSTRACT: External beam proton-induced X-ray emission (PIXE) analysis has been used to verify the presence of lead in the finger bone of a murder victim. The deceased, who had been buried several years, was known to have suffered a bullet wound to his right hand several years before death. X-ray radiographs of the right second proximal phalanx revealed the possible presence of metal fragments below the surface of the bone. To verify the presence of lead in a nondestructive manner, the bone was scanned with a 1.5-MeV proton beam. PIXE analysis showed that lead was present only in the vicinity of the fragments previously detected in the radiographs. A study of gunshot residue in bone shows that the distribution of lead around the bullet hole is independent of the firing distance for distances greater than 0.6 m.

KEYWORDS: physical anthropology, ballistics, X-ray analysis, gunshot residues

We have employed proton-induced X-ray emission (PIXE) analysis to identify lead residue from a bullet in a bone in a case where the point of impact of the bullet on the bone has been obscured. The investigation was initiated by the need to identify the presence of lead in the finger bone of a murder victim using a nondestructive method. The specimen analyzed was from a skeleton unearthed in an isolated grave in Rhode Island in 1983. The skull exhibited a through-and-through transverse gunshot wound of the cranial vault. The presumed decedent was a 22-year-old white male who had disappeared from Providence, RI in 1975. Dental records were not available; however, two years before his disappearance, the presumed decedent had been admitted to a local hospital with a gunshot wound in the right hand. X-rays taken at the time of his admission had been destroyed but the clinical records described a small-caliber bullet wound resulting in an open fracture of the second right metacarpal. The record states that "There was a 5 mm entrance wound at the base of the second metacarpal and an exit wound over the dorsum of the proximal phalanx approximately 1 cm in diameter . . . ."

Gross examination of the bones of the right hand of the skeleton revealed a healed traumatic exostosis of the second metacarpal bone. Radiographs showed metallic fragments em-

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bedded in the cortical bone of the second right proximal phalanx (Fig. 1) approximately 1 mm from the surface of the bone. These fragments ranged from about 0.1 to 3 mm in greatest diameter. Positive identification of the decedent required a correlation of the clinical records with the metallic fragments in the proximal phalanx. Proton-induced X-ray analysis was undertaken to determine whether or not the fragments observed in the radiographs were lead.

Method

The use of characteristic X-rays to detect trace elements is a well established analytical technique. Conventional X-ray analysis such as X-ray fluorescence (XRF) uses an intense X-ray beam to excite X-ray fluorescence in the sample. Alternatively, a focused electron beam is used in a scanning electron microscope (SEM-XRF) or electron microprobe (EMP) to excite the characteristic X-rays. In recent years, the use of ion beams as a source of excitation has proven to be a complimentary method to XRF. Proton-induced X-ray (PIXE) analysis [*1*] utilizes a beam of energetic (1- to 2-MeV) protons to induce inner-shell ionization near the surface of the sample. Subsequent to ionization, the atom emits one of several characteristic X-rays, the energy of which identifies the element. The number of characteristic X-rays (that is, the intensity) is a measure of the elemental concentration in the sample. Using a multichannel analyzer and a nondispersive X-ray detector with high energy resolution to accumulate the proton-induced X-ray spectrum of all elements heavier than sodium, the concentration of these elements can be determined.

The experimental arrangement is shown schematically in Fig. 2. The proton beam from a 3-MeV Van de Graaff accelerator exits from the accelerator beam line through a thin win-

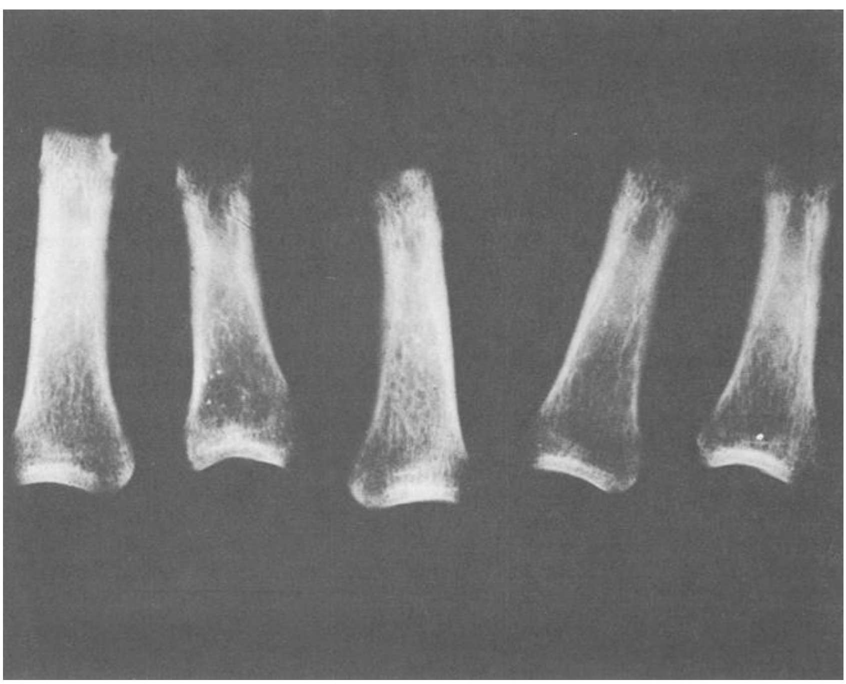

FIG. 1—*Radiograph of victim's phalanges. Note the fragments in the second bone from the left in the radiograph.*

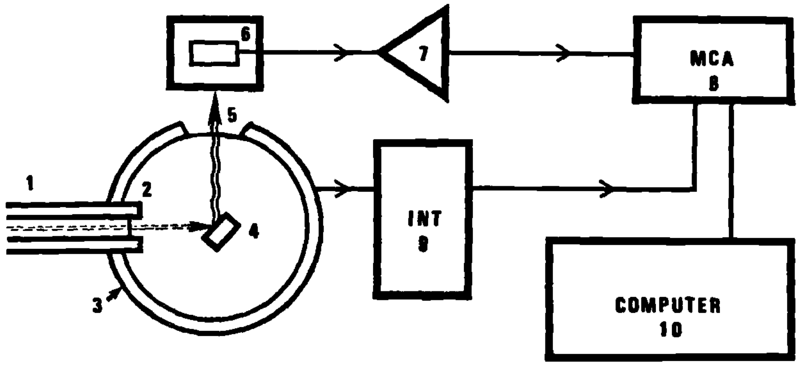

FIG. 2—*Schematic of the experimental arrangement:* (1) *accelerator beam line,* (2) *exit window,* (3) *sample chamber,* (4) *sample,* (5) *capton window,* (6) *X-ray detector,* (7) *amplifier,* (8) *multichannel analyzer,* (9) *current integrator, and* (10) *computer.*

dow into a sample chamber filled with helium gas at 1-atm pressure. For this investigation, the bone samples were mounted on a micromanipulator so that they could be scanned with a 2-mm diameter proton beam which determined the spatial resolution of the elemental concentration. Because the X-ray yield is proportional to the number of protons incident on the sample, the X-ray intensity is scaled by the total charge incident on the sample. The current of the 1.5-MeV proton beam was 7 nA.

PIXE analysis using a proton beam external to the accelerator beam line offers advantages over other X-ray analysis methods.

1. The analysis of large[4] and odd shaped objects is possible without special sample preparation. The object to be analyzed does not have to be introduced into a vacuum.
2. Samples can be scanned, thus allowing localized trace element analysis. The diameter of the proton beam can be adjusted from several millimetres to several micrometres.
3. The sensitivity is approximately an order of magnitude better than electron-induced X-ray fluorescence (EMP or SEM-XRF) and, in some instances, better than photon-induced XRF.

Results and Discussion

The presumed decedent's second right proximal phalanx was mounted in front of the proton beam and scanned in a horizontal and lateral direction. A strong X-ray peak characteristic of lead was observed in a lateral scan approximately 3 cm distal to the proximal end. The proton-induced X-ray spectrum obtained at this position (Fig. 3) displays lead L X-ray lines at 10.6 and 12.6 keV. The high sensitivity of the PIXE analysis is indicated by the observation of the trace elements zinc, copper, manganese, and potassium which are present in human bone at the 10 to 100 part per million level.

Figure 4 shows the lead X-ray intensity as a function of lateral position at the height indicated by the arrow. The maximum intensity occurs in the area where the presence of lead would be expected according to the radiograph. The bone was rotated 180° from the position of the maximum; however, no trace of the lead was detected on the back side. Several other phalanges of the victim were scanned in the same manner but no lead was found.

[4]The PIXE system at the University of Oklahoma presently allows the mounting of objects about 10 cm in length and about 5 cm in diameter. The use of the University's XRF facility would have required that the victim's phalanx be cut in order to fit the sample chamber. However, since the bone was to be produced as evidence in a trial, it was important to keep the bone intact.

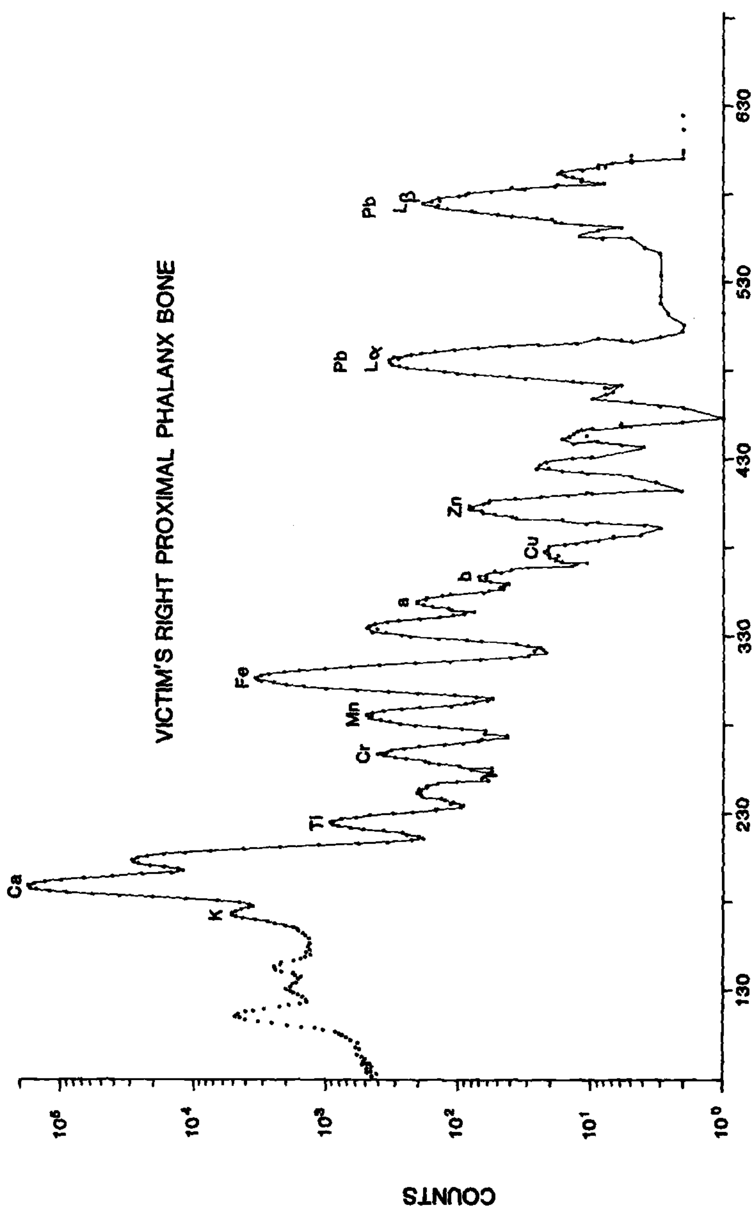

FIG. 3—*Proton-induced X-ray spectrum of the victim's second right proximal phalanx bone indicating the presence of lead.*

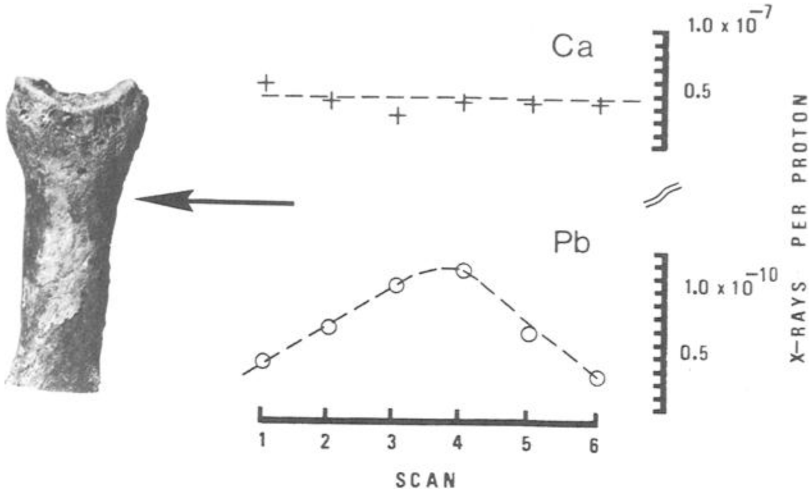

FIG. 4—*Lateral scan of the lead and calcium concentration of the victim's right proximal phalanx bone 3 cm distal from the proximal end of the bone.*

As PIXE typically yields elemental concentrations in a micron layer at the surface of the sample, the lead detected here is probably residue left by the bullet near the surface of the bone rather than the fragments seen in the radiograph which are 1 mm or more below the bone surface. However, given the pitted condition of the bone, it is possible that some of the fragments in the radiograph were directly exposed to the proton beam.

Sen et al [2] have used the PIXE technique to study gunshot residue profiles near bullet holes. They prepared gunshot residue samples in the laboratory by firing bullets through filter paper from various distances. As might be expected, they found that the relative intensity of the residue elements, including lead, decreased as the distance from the bullet hole increased. Furthermore, as the distance between the gun and filter paper increased, the radial distribution of the residue decreased from 27 mm at a firing distance of 15 cm to a few millimetres at a firing distance of 76 cm.

We have measured the radial distribution of lead around bullet holes in the pelvic bone of a cow produced by firing a .22-cal bullet from distances between 0.6 and 3 m. The bullet passed completely through these test bones. The results (Fig. 5) show that at distances between 0.6 and 3 m, the radial distribution is independent of the firing distance. The scatter of the data points reflects the ragged edge of the bullet hole in the bone. The distribution decreased exponentially from the edge of the hole with a half width of about 1 mm. The amount of lead residue at the edge of the hole was found to be about $5 \cdot 10^{17}$ atom/cm^2. This is of the same order of magnitude as the lead concentration observed at the peak of the lateral scan of the victim's right proximal phalanx shown in Fig. 4. This observation supports the hypothesis that the lead observed in the scan is residue left by the bullet fragments passing through the bone surface rather than the direct observation of the fragments shown in the radiograph which are embedded in the bone at a depth greater than the estimated range of the proton beam. The lead distribution can therefore be used to identify the location of the bullet entry in cases where this can not be done by visual inspection.

We have investigated whether charring the bone would leave the lead residue intact. For this study, a bone with a bullet hole was heated above the melting point of lead to about 370°C for 10 min leaving the bone charred and very brittle but still solid enough to be mounted in front of the beam. No significant difference between the lead concentration before and after heating was found. In addition, we investigated whether the presence of tissue

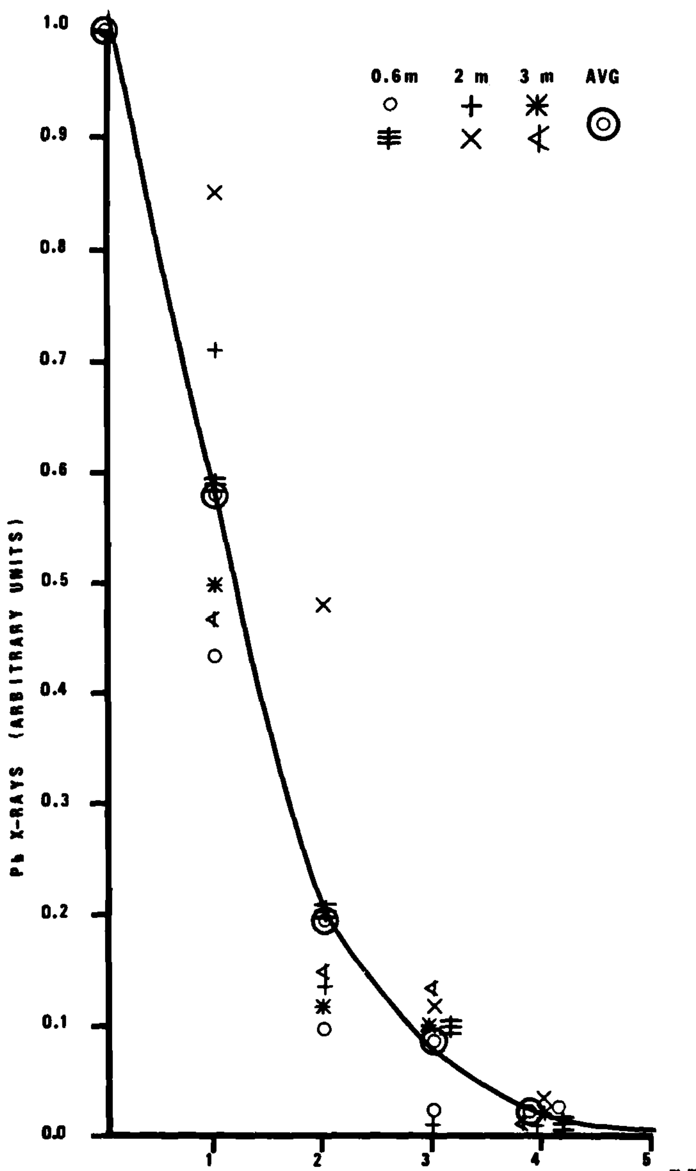

FIG. 5—*Relative concentration of lead near the surface of the bone as a function of distance from the edge of the bullet hole at firing distances of 0.6, 2, and 3 m.*

on the bone would affect the distribution of lead. A .22-cal unjacketed rimfire bullet was fired from a Remington automatic rifle from a distance of approximately 2 m into a bone covered with tissue which was subsequently removed. A comparison of this bone to other nontissue covered bones showed no significant difference in the lead distribution around the bullet hole.

Summary

Proton-induced X-ray analysis has been used to detect lead residue caused by the passage of a bullet through a bone. The presence of tissue covering the bone does not appear to affect significantly the distribution of lead residue around the bullet hole. At distances of 0.6 m or more this distribution appears to be independent of the distance from which the bullet was fired. Charring the bone at temperatures as high as 370°C does not affect the detection of the lead residue. The ease of sample preparation and the capability to analyze large objects nondestructively make PIXE an attractive alternative to other trace element analysis methods if an accelerator facility is available.

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