

Portable X-Ray Fluorescence (pXRF) Analysis of Heavy Metal Contamination in Graveyards with Contrasting Soil Types

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Abstract

Human remains have been interred in burial grounds since historic times. Although the re-use of graveyards differs from one country, region or time-period to another, over time graveyard soil may become contaminated or enriched with heavy metal elements. This paper aims to present heavy metal element soil analysis from two UK church graveyard study sites with contrasting necrosols, but similar burial densities and known burial ages dating back to the 16th Century and some possibly older than 1,000 years. Portable X-Ray Fluorescence (pXRF) element analyses were undertaken, both *in-situ* on surface locations and laboratory-based on surface and near-surface soil pellets. Results show elevated levels of Pb, Mn, Cr, Cu, Zn and Ca in both necrosols when compared to background values. Element concentration anomalies remained consistently higher than background samples down to 2 m bgl, where sampled, but reduced away from church buildings which may reflect burial densities. Element concentration anomalies were higher in the clay-rich necrosol than in sandy necrosol. Field-based rapid measurements gave similar relative concentration values to laboratory-based soil pellet measurements, although laboratory-based analyses were more precise. Study results implications suggest that long-used necrosols are likely to be more contaminated with heavy-metal elements than similar soil outside graveyards with implications for burial grounds management, adjacent populations and where burial grounds have been deconsecrated and turned to residential dwellings.

Introduction

All burial grounds are unique both in their natural environment (including soil type, parent material, vegetation, topography and climate) as well as anthropogenic burial numbers/styles/depths (Fig. 1), body distributions and above-ground placement of memorials and installation of pathways and roads to access the site. The mechanical disturbance via re-excavation and re-infilling of burial sites, alongside varying above ground vegetation types, as well as the presence of human remains, make graveyard soils unique and have their own soil type category; necrosols (Amuno and Amuno, 2014; Asare et al., 2020). Necrosols, despite increasing numbers of cemeteries and burial sites both in rural and urban environments, are poorly understood, especially regarding potential contamination and ecological risks (Jonker and Oliver, 2012). This is largely due to the complex biological and chemical processes occurring in these soils, resulting in both spatial and temporal heterogeneity of necrosols (Amuno and Amuno, 2014).

Reuse of graveyard and cemetery sites for burying human remains has been happening for at least 10,000 years since Early Mesolithic times (Schulting et al., 2019). The practice of reusing existing graveyards differs by country, region and timescales of clearing old graves before new ones are emplaced. For example, the United States generally leave human remains untouched *in situ* in perpetuity, whereas in the United Kingdom it is common to have a 100-year period, by which time any direct relatives should have died before an existing graveyard can be reused (Mytum, 2000), and in Germany remains can be moved when only buried for 25 years and a fresh grave emplaced (Fiedler et al., 2009).

Soil from graveyard sites differ from the natural soil profile largely through disturbance and due to the nature of the material buried. Previous research have likened graveyards to landfills (Fiedler et al., 2012), with elevated levels of organic matter (Kim et al., 2008), embalming fluids (Chiappelli and Chiappelli, 2008; Uslu et al., 2009), creosote from coffins (Mininni et al., 2007), as well as materials from the bodies themselves (Fiedler et al., 2012). In a few cases, cemetery materials carried in soil water have also been found to have contaminated local groundwater supplies with pathogens, viruses and heavy metals (Knoefes and McGee, 2002; Matias et al., 2004; Kim et al., 2008).

Non-invasive geophysical studies in such burial grounds indicate elevated conductivity levels in grave soil (Hansen et al., 2014), with individual grave geophysical anomalies decreasing with increasing burial age, when compared to background values. However, soil texture and moisture content have been shown to be major variables with sandy soils causing leaching of grave contents well beyond the grave-cut, whereas clay-rich soils tend to retain these fluids within the grave-cut itself (Pringle et al., 2012, 2016; Dick et al., 2017).

Archaeological studies have shown ancient burial ground soils to have elevated levels of heavy-metal elements such as Iron, Lead, Manganese and Copper (Amuno and Amuno, 2012, Jonker and Oliver, 2012), as well as other elements such as Phosphorus and Nitrogen (Bethell and Carver, 1987; Asare et al., 2020), with human exposure to such toxic metals causing kidney damage (Khan et al., 2011) and links to Parkinson's and Alzheimer's disease (Mohod and Dhote, 2013). This may have important health implications for local graveyard workers, residents in the surrounding areas as well as potential risks to the local environment via leaching into surrounding soils and groundwater (Jonker and Oliver, 2012). However, there has been, to-date, limited research on the soil contamination potential of cemeteries and graveyards

X-ray fluorescence spectrometry (XRF) is an analytical technique for the chemical characterisation of environmental materials. Although traditional, laboratory-based, wavelength dispersive XRF spectrometry will undoubtedly remain the analytical method of choice for studies in which data must be of the highest possible quality, such analyses inevitably involve greater costs, lengthier sample preparation and data processing procedures, and longer analysis times.

Portable XRF (pXRF) field surveys have been shown to be effective for rapid evaluation of heavy metal soil contamination (Radu and Diamond, 2009; Brent et al., 2016; Rouillon et al., 2017; Liang et al., 2018), biogeochemical mapping over mine tailings (Rincheval et al., 2019),

archaeological object studies (Kasztovsky et al., 2018; Michalowski et al., 2020), marine microplastics (Turner, 2017), species profiling (Nganvongpanit et al., 2016), and even lead levels in living human bones (Zhang et al., 2021). The ability to carry out *in-situ* analysis for many other elements means that the methodology has considerable application as a field-based analysis tool (Frahm and Doonan, 2013).

However, soil water content is highly naturally highly variable across different land uses, soil types and habitats, which can be somewhat challenging when conducting physical measurements for site comparisons, for example, electrical resistivity surveys (Jervis and Pringle, 2014). This holds true for pXRF analysis, with the additional complication that the attenuation of X-rays by water is a function of the energy used to characterize the elements of interest. Low-energy X-rays are more strongly attenuated than high-energy ones, thus for pXRF analysis, elements with lower atomic masses are more strongly impacted. Studies have shown that for every 1% increase in soil water content, there is a 1.15% – 1.75% decrease in reported elemental concentration for Mn through to As, while elements lighter than Mn are even more greatly attenuated (Parsons et al., 2013; Imanishi et al., 2010). This means that sample water content must be measured and corrected for, as light element values determined *in situ* will likely not be directly comparable to results of a traditional laboratory analysis (Padilla et al., 2019).

The two aims of this study were therefore: (1) to determine the utility of pXRF measurements by comparing field- and lab-based pXRF measurements both spatially and temporally (down the soil profile) and; (2) to evaluate the potential heavy metal contamination of long-used (500+ years) church necrosol graveyards with different soil textures. Study objectives were therefore to: (1) collect pXRF field surface graveyard soil measurements and then take samples for subsequent laboratory soil pellet analysis, (2) collect surface samples and shallow surface soil auger profiles in long-established church graveyards for pXRF laboratory sample analysis in sandy and clay soil types respectively, (3) assess the respective known burial population and areal extent to quantify the respective graveyard burial histories and, (4) analyse pXRF results, compare to other studies and suggest implications for those living adjacent to burial grounds, adjacent to water supplies and re-use of ex-burial grounds.

Materials And Methods

Graveyard sites background

Two Church of England graveyards were selected for this study, study site 1 at St. John's Church in Keele Staffordshire, UK (Fig. 2), and study site 2 at St. Michael and All Angels' Church in Stockton, Norfolk, UK (Fig. 3). The reason these two were studied is that they have over known burial records from over 500 years to the present day, contrasting soil textures (site 1 is sandy and site 2 is clay-rich soils respectively), bedrock types, rainfall levels and geographic settings (semi-urban and rural respectively) which make a useful contrast for this study (Dick et al., 2017).

Study site 1 is located in Keele Village in Staffordshire, situated ~200 m above sea level and with an average rainfall of 806 mm/yr. The site hosted a Knights Templar church built around 1160 CE, before being taken over by the Knights Hospitaller in 1324 CE. A new church was built by the Sneyd family in the 16th Century, before the present sandstone church was built in 1868-1870 CE (Pevsner, 1974). A desktop study, confirmed by soil auger reconnaissance (Fig. 4a) showed there to be a sandy loam soil overlying the Upper Carboniferous Butterton Sandstone Member of the Halesowen Formation bedrock found at ~2.5m bgl.

Study site 2 is located in Stockton Village in Norfolk, situated ~35 m above sea level, with a lower average rainfall of 620 mm/yr. The church of St. Michael and All Angels is present on the study site, with a Saxo-Norman-style round tower, probably dating from the 13th – 14th Century, and with later medieval flintwork additions (Knott, 2005). A desktop study, confirmed by soil auger reconnaissance (Fig. 4b), indicate that the study site is located on clay-rich soil, derived from a glacial diamicton, overlying clays and sands of the Pleistocene Beccles Formation found at ~2.5m bgl.

Known burial Records

For site 1 at St. Johns, available graveyard burial records of 5,735 individuals were available from 1585-1970 CE and 1990–2018 CE (see Fig. 5a and Supplemental Data), the data gap due a records office fire, averaging 13/year, with a typical increase after the Industrial Revolution (the church being situated on the edge of the industrialised area of Stoke-on-Trent), before declining in the 20th Century.

For site 2 at St. Michaels, available graveyard burial records of 669 individuals were available from 1561–2018 CE (see Fig. 5b and Supplemental Data), averaging 1/year, with a typical increase and decrease in burial rate with the Industrial Revolution and the 20th Century, respectively, as observed in case study 1.

When known burial numbers were corrected for respective graveyard areal extents (10,800 m² for study site 1 at St. John's; and 2,330 m² for study site 2 at St. Michaels), similar burial densities of ~2.2 individuals/m² at study site 1 and ~3 individuals/m² at study site 2 were determined to be approximately comparable.

Both graveyards had mostly marked earth-cut graves with headstones, with burial ages that range from the 18th century to the present day (Fig. 5). At study site 1, there were more multiple familial burials in the same grave which results in more variable coffin burial depths. A general review by Hart and Casper (2004) found average depths (bgl) of 1.4 m for one coffin, 1.83 m for two coffins and 2.7 m for four coffins for familial burials.

Soil sampling and analysis

Research on optimal sampling methodologies has suggested at least five samples should be acquired in different locations on a site to gain representative results (Pye et al., 2005, 2007; McKinley and Ruffell, 2007). Field reconnaissance determined sampling areas (for example, avoiding areas of dense vegetation/trees, etc., which prohibited sampling), from which the 77 and 64 specific sample positions were selected (via a random generator to avoid potential sampling bias) within 5 m x 5 m grids at study site 1 St John's and study site 2 St. Michael's Churches respectively. Two additional control samples in the same soil type were also taken ~100 m away from each graveyard to act as a control.

In-situ versus laboratory pXRF surface soil sampling

First, to compare pXRF surface measurements of *in-situ* with laboratory soil samples, overlying vegetation was firstly removed to expose the soil at each sampling location, which was then checked for pebbles or vegetation which would contaminate the measurement (Fig. 6a). The soil was then analysed with a hand-held pXRF instrument for a two-minute measurement period, since initial tests of one soil pellet sample indicated a 120 second field sample time would give acceptable two-sigma errors (determined by the instruments internal software via counting statistics) of up to $\pm 20\%$ error (Pringle et al. in press FSI). Elemental analysis was undertaken with a ThermoScientific™ NITON XL3t 900 Analyser with in-built factory calibration (Fig. 6b). The instrument employs four sub-filters (Main, Low, High, Light), each targeting a specific range of elements. Each of these filters was allotted an equal proportion of the overall analytical time. Analytical precision and accuracy were evaluated using a randomly selected, unprepared soil sample from study site 1 St. John's Church, and a range of international standard reference materials (NIST 2780, BHVO-1, AC-E, AGV-1). Repeat measurements of hand-held analyses indicated precision (expressed as relative two-sigma standard deviation) within 11% for Zinc, 7% for Manganese, 4% for Lead and 25% for Chromium. Accuracy during hand-held analysis was found to be affected variably by the method for the elements given above, with deviation from known values ranging from 23–93%.

For the pXRF laboratory soil measurements, once the *in situ* measurements had been collected, ~250 g of this surface soil from the top 5 cm was collected at sampling positions in both study sites, bagged, labelled and stored at 4° C. A subset of samples was also taken for routine soil characterisation analysis, initially including determination of average electrical conductivity following standard methodologies.

Due to the laboratory soil pellet preparation time, 33 of out of the 78 *in situ* soil samples from study site 1 and 31 from 64 surface soil samples from study site 2 graveyards were oven-dried for 24 hours at 105°C before being hand-ground using a pestle and mortar to pass through a 63 µm sieve. The sample was then mixed with 1.5 mL of polyvinylpyrrolidone-methylcellulose binding agent and mechanically pressed under 10 Tn pressure (Fig. 6e) into a homogenous flat-cylinder pellet, before being oven dried for a further 24 hours at 105°C to remove any soil water influence on signal strength (Kalnicky and Singhvi, 2001). Each pellet (Fig. 6f) was then placed over the fixed aperture of the NITON XL3t 900 Analyzer in its laboratory holder and analysed using a five minute measurement time as there was not a time restriction on measurement time unlike the *in situ* field measurements. Laboratory-based analyses typically yielded precision (expressed as relative two-sigma standard deviation) within 7% for Zinc, 6% for Manganese, 4% for Lead and 16% for Chromium. Typical accuracy (expressed as average percentage deviation from known values) for laboratory analyses was within 8% for Zinc, 3% for Manganese and 3% for Pb.

Shallow surface soil auger profiles

Secondly, to collect shallow-surface (down to 0.75m bgl) soil samples of the two study sites, adjacent to 32 and 31 of the sampling locations at study site 1 and study site 2 graveyards respectively, a 0.75m slimline hand auger was used with a hammer to hand-extract three (n=3) ~250g of soil samples at 0m - 0.25m, 0.26m - 0.5m and 0.51m - 0.75m soil depth bgl ranges (Fig. 6c). The resulting soil samples were then labelled and stored in polyethylene bags at 4°C until soil pellets from each of the listed soil auger profile depth ranges were generated with the procedure as already detailed and then each pXRF analysed in the laboratory for a 5 minute measurement time as previously described.

Empty grave soil wall profile *Thirdly*, we were able to collect deeper subsoil elemental data, since a grave was being hand excavated at study site 1. Here, we collected ~250g of soil samples every 0.25 m from 0 m to 2 m bgl on the south end of the empty grave (Fig. 6d). The resulting soil samples were labelled and stored in polyethylene bags at 4°C until soil pellets were generated and pXRF analysed in the laboratory for a 5 minute measurement time procedure as already detailed.

Results

Basic soil characterisation

The field-collected samples had routine soil characterisation analysis, including determination of electrical conductivity, pH and soil moisture content following standard methodologies. Study site 1 electrical conductivity recorded an average of 47 $\mu\text{S}/\text{cm}$ (28 SD), with study site 2 recorded an average of 99 $\mu\text{S}/\text{cm}$ (49 SD). Study site 1 ph recorded an average of 6.3 (0.8 SD), with study site 2 recorded an average 7.8 (0.8 SD). Study site 1 soil moisture content ranged from 8% - 27% (14.1%), with study site 2 soil moisture content ranged from 13% - 27% (average 21%).

In-situ versus laboratory pXRF surface soil sampling

Our results show clear differences in element concentrations in the soil samples measured, both within each graveyard and between the two graveyards.

At study site 1, the sand-rich soil graveyard study site, *in-situ* surface fresh soil pXRF measurement results showed lead element concentrations varying from 21 mg/kg up to a maximum of 530 mg/kg with an average of 121 mg/kg (Table 1), with relatively higher concentrations adjacent to the church itself (Fig. 7). Manganese had also high concentrations, varying from 210 mg/kg up to a maximum of 1,910 mg/kg with an average of 564 mg/kg (Table 1) but the distribution across the graveyard was different, when compared to lead distributions. Arsenic had relatively low concentrations, varying from 6 mg/kg up to 26 mg/kg with an average of 10 mg/kg, the same concentration as observed from the control soil samples (Table 1). Zinc had concentrations varying from 22 mg/kg up to 241 mg/kg and averaging 60 mg/kg, interestingly 10 mg/kg less than the 70 mg/kg average observed from the control samples (Table 1). Chromium had low concentrations, compared to 99 mg/kg from control soil samples, of 15 mg/kg up to 55 mg/kg and average of 31 mg/kg. By contrast, the soil pelletised laboratory pXRF measurement results from the same locations showed consistently higher heavy metal concentrations when compared to the *in-situ* measurements (Fig. 7 and Table 2), with lead levels varying from 132 mg/kg up to 1,541 mg/kg, but the relative element concentration areas were similar.

Table 1

Descriptive statistics of heavy metal element pXRF concentration *in-situ* surface soil and laboratory soil pellets over 2 min and 5 min measurement duration respectively, acquired from study site 1. Raw data in Supplementary Material. Av = average, n = number of analyses

Element	<i>In situ</i> surface (n=77) fresh soil measurements (mg/kg)				Laboratory (n=32) dry soil pellets (mg/kg)				Control soil (n=2) dry soil sample pellets (mg/kg)
	Min.	Av.	Max.	SD	Min.	Av.	Max.	SD	Av.
Pb	21	121	530	96	132	381	1541	331	51
As	6	10	26	6	6	11	25	4	10
Mn	210	564	1910	387	107	1629	3335	669	1104
Zn	22	60	241	37	42	125	384	77	70
Cr	15	31	55	10	38	70	155	23	99

Table 2

Descriptive statistics of heavy metal element pXRF concentration *in-situ* surface soil and laboratory soil pellets over 2 min and 5 min measurement duration respectively, acquired from study site 2. Raw data in Supplementary Material. Av = average, n = number of analyses

Element	<i>In situ</i> (n=64) surface fresh soil measurements (mg/kg)				Laboratory (n=31) dry soil pellets (mg/kg)				Control soil (n=1) dry soil sample pellets (mg/kg)
	Min.	Av.	Max.	SD	Min.	Av.	Max.	SD	Av.
Pb	11	246	6604	851	70	582	4242	867	28
As	4	8	26	5	4	11	29	5	8
Mn	110	192	320	82	247	527	1562	288	486
Zn	23	195	3007	433	72	604	7408	1391	95
Cr	16	30	51	8	10	67	97	22	36

At study site 2, the clay-rich soil graveyard study site, the *in-situ* surface soil pXRF measurements showed very high lead concentrations, varying from 11 mg/kg up to a maximum of 6,604 mg/kg with an average of 246 mg/kg (Table 1), again with higher concentrations measured adjacent to the church (Fig. 8). Zinc had also high concentrations, varying from 23 mg/kg up to 3,007 mg/kg, with an average of 195 mg/kg (Table 2) but the distribution across the graveyard was more varied when compared to the lead distributions. Arsenic had similar concentrations as background control samples, varying from 4 mg/kg up to 26 mg/kg with an average of 8 mg/kg. Manganese had relatively low concentrations,

when compared to background control samples, varying from 110 mg/kg up to 320 mg/kg with an average of 192 mg/kg. Chromium had similar concentrations to background control samples, varying from 16 mg/kg to 51 mg/kg with an average of 30 mg/kg. By contrast, the study site 2 soil pelletised laboratory pXRF measurements from the same locations showed consistently higher heavy metal concentrations, when compared to the *in-situ* measurements (Fig. 7 and Table 2), other than Arsenic and chromium. For example, lead levels varied from 70 mg/kg up to 4,242 mg/kg with an average of 582 mg/kg, but the relative element concentrations between elements were again similar.

Graphical cross-plots of lead concentrations from *in-situ* versus laboratory measurements show a good correlation for study site 2 clay soils, but a poor correlation for study site 1 sandy soils (Fig. 9), suggesting elements are retained in the clay soil but perhaps leached in sandy soils.

The variation between *in-situ* fresh surface and soil pellet analyses was most probably due to both a consistent flat measuring surface in the soil pellets when compared to the *in-situ* surface measurements, and the removal of water in the laboratory samples, but the relative differences in element concentrations within the graveyard were found to be similar with both analysis methods. Basic descriptive statistics evidenced for study site 1 sandy soil an average of 120 mg/kg for *in situ* surface soil lead concentrations and 215 mg/kg for soil pelletised lead concentrations, a 230% difference, and for study site 2 clay soil an average of 121 mg/kg for *in situ* surface soil lead concentrations but 582 mg/kg for soil pelletised lead concentrations, a 590% difference (Tables 1-2).

As soil sample locations were geospatially referenced, elements could be compared directly with the analysis of distance from the respective churches at each study site (Fig. 10). All elements showed decreasing concentrations trends with increasing distance from the church buildings but these were not statistically significant. Interestingly, the calcium levels were recorded at very high concentrations adjacent to both church buildings (>10,000 mg/kg) when compared to ~1,000 mg/kg average control values (Fig. 10).

Shallow surface soil auger profiles

At study site 1, the sand-rich soil study site, most of the heavy element concentrations reduce with increasing soil depth bgl, for example Manganese concentration averages increasing from 1937 mg/kg at 0-0.25m, 2351 mg/kg at 0.26-0.5m and 2039 mg/kg at 0.51-0.75 m bgl which contrast with 1115 mg/kg for the control samples. Lead concentrations were consistently high, averaging 650 mg/kg at 0-0.25m, 897 mg/kg at 0.26-0.5m and 785 mg/kg at 0.51-0.75m bgl (Fig. 11), which contrasts with 25 mg/kg control levels, although arsenic levels were at relatively low levels averaging 7 mg/kg to 18 mg/kg (Table 3).

Table 3
Descriptive heavy metal element pXRF concentration statistics of laboratory soil depth range pellets over 5 min measurement duration respectively, acquired from study site 1 sandy soil. Av = average, n = number of analyses

Selected Element	0-0.25m bgl (n=15) dry soil pellets				0.26-0.5m bgl (n=15) dry soil pellets				0.51-0.75m bgl (n=15) dry soil pellets							
	Graveyard (n=15)				Control (n=2)				Graveyard (n=15)				Control (n=2)			
	Min.	Av.	Max.	SD	Av.	Min.	Av.	Max.	SD	Av.	Min.	Av.	Max.	SD	Av.	
Pb	116	650	2,575	684	74	49	897	5962	1612	54	6	785	4959	1437	25	
As	5	19	32	8	12	-	11	35	11	10	-	7	20	8	9	
Mn	763	1937	3145	677	952	452	2351	3772	1057	1245	354	2039	3109	883	1115	
Zn	44	113	217	44	82	46	94	204	43	71	18	70	131	27	56	
Cr	64	127	207	38	96	61	158	242	53	102	52	132	225	55	99	

At study site 2, the clay-rich soil study site, all selected heavy element concentrations increase with increasing soil depth bgl, with lead levels in particular increasing, averaging from 70 mg/kg at 0-0.25m, 130 mg/kg at 0.26-0.5m to 1078 mg/kg at 0.51-0.75 m bgl (Fig. 11), which contrasts with the 53 mg/kg control sample levels. Zinc concentration levels were also high at 108 mg/kg at 0-0.25m, 125 mg/kg at 0.26-0.5m and 1457 mg/kg at 0.51-0.75m bgl (Fig. 11), which contrasts with the 138 mg/kg control sample levels. Arsenic levels were also at low and similar concentrations to the control average of 6 mg/kg - 10 mg/kg (Table 4).

Table 4

Descriptive heavy metal element pXRF concentration statistics of laboratory soil depth range pellets over 5 min measurement duration respectively, acquired from study site 2 clay-rich soil. Av = average, n = number of analyses

Selected Element	0-0.25m bgl (n=6) dry soil pellets (mg/kg)					0.26-0.5m bgl (n=6) dry soil pellets (mg/kg)					0.51-0.75m bgl (n=6) dry soil pellets (mg/kg)				
	Graveyard (n=7)			Control (n=1)		Graveyard (n=7)			Control (n=1)		Graveyard (n=7)			Control (n=1)	
	Min.	Av.	Max.	SD	-	Min.	Av.	Max.	SD	-	Min.	Av.	Max.	SD	-
Pb	27	70	179	56	17	42	130	385	121	26	83	1078	4830	2099	53
As	6	10	12	3	8	7	9	11	2	9	4	8	10	2	6
Mn	382	495	588	78	499	194	388	570	144	457	288	412	591	113	502
Zn	54	108	303	97	68	63	125	349	110	79	91	389	1457	598	138
Cr	53	105	285	89	46	35	71	106	25	29	41	58	72	11	33

Empty grave soil wall profile

Here, although we found that the cross-plot concentrations versus depth bgl correlations were not statistically significant ($P > 0.05$), there were still trends of element concentrations reducing with increasing soil depth, for example lead reducing from 1591 mg/kg at the surface down to 628 mg/kg at 2m bgl, apart from chromium which did not show this trend (Fig. 12). The calcium concentrations reduce with depth back to 500 mg/kg, slightly higher than the national average of 400 mg/kg for sandy rural soils as given by Ross et al. (2007).

These results (Table 5) contrast with the over lying shallower depths lead concentration results which do not show much reduction of lead concentrations with depth up to 0.75m bgl but note this is only one empty grave sample at one location within the graveyard.

Table 5

Descriptive heavy metal element pXRF concentration statistics of laboratory soil depth range pellets over 5 min measurement duration respectively, acquired from study site 2 St. Michael and All Angels graveyard, Stockton, Norfolk, UK.

Depth (m)	Selected heavy metal element concentrations (mg/kg)					Ca (mg/kg)
	Pb	As	Mn	Zn	Cr	
0.0	1591	11	2213	247	74	2465
0.25	808	11	1501	360	109	1765
0.5	1447	-	1530	335	71	2067
0.75	1070	5	1138	305	49	1630
1	966	6	1113	345	86	1826
1.25	593	7	953	310	85	1271
1.5	615	5	1442	255	65	737
1.75	561	6	1662	253	72	752
2.0	628	6	1905	305	78	760

Discussion

Addressing the first aim of this study, to determine the utility of pXRF measurements and comparing to laboratory measurements, analysis of both *in situ* surface soil samples and subsequent laboratory analysis of soil pellets at both sandy- and clay-rich soil church graveyard study sites have shown significant heavy metal element contamination. The laboratory measurements of soil pellets derived from samples taken at the surface gave higher element concentrations, when compared to the *in situ* pXRF surface measurements. On average, measured element surface:lab concentration ratios were surprisingly similar at 1:2.3 and 1:2.4 for case study 1 sandy soil and case study 2 clay soil graveyards respectively. This was most probably due to the removal of soil moisture in the laboratory samples and having more flat, consistent measuring surfaces (Tables 1 and 2).

However, both datasets still gave relatively similar relative concentration variations across the study sites (Figs. 5 and 6). Moreover, the concentrations determined by both field and laboratory methods for lead, the principal element of interest in the churchyard case study, were well above the control averages (381 mg/kg and 582 mg/kg averages for case study sites 1 and 2 respectively compared to 51/28 mg/kg control) and are well above the typical background concentrations observed in the soils of England and Wales (i.e. 52 mg/kg taken from Ross et al. 2007). Similarly zinc concentrations were also relatively high, well above control averages (125 mg/kg and 605 mg/kg averages for study sites 1 and 2 respectively compared to 70/95 mg/kg control) and are also above the typical background concentrations observed in the soils of England and Wales (i.e. 81 mg/kg taken from Ross et al. 2007) – see Table 6.

Table 6

Summary statistics of this study surface soil pellet pXRF results compared to other studies and *mean UK soil values from Ross et al. (2017)

Element	Case study 1 sandy soil av. pellets (mg/kg)	Case study 1 control sandy soil (mg/kg)	Case study 2 clay soil av. pellets (mg/kg)	Case study 2 control clay soil (mg/kg)	Neckel et al. (2016) 3 cemeteries av. surface grave soil (mg/kg)	Fiedler et al. (2012) coffin material (mg/kg)	*Mean UK rural soil (mg/kg)
Pb	381	51	582	28	34	492	52.5
As	11	10	11	8	-	4.6	10.9
Mn	1629	1104	527	486	304	-	612
Zn	125	70	604	95	105	821	81.2
Cr	70	99	67	36	25	26.2	34.4

Therefore, these results indicated a high level of heavy metal element contamination exists at these long-lived graveyards and reinforces the Fiedler et al. (2012) study that graveyards are effectively a variety of landfill. This study showed that rapid *in situ* surface analysis techniques can indeed identify priority areas for subsequent follow-up urgent inspection which reinforces Young et al. (2016) with a similar study on rock materials. If there were nearby surface and groundwater supplies for the local human population then potential heavy metal contamination from these long-used necrosol graveyards would clearly be of some concern, as detailed by Oliveira et al. (2012) and Matias et al. (2004) evidenced from a Portuguese graveyard study and nearby water borehole results.

With respect to the second aim, to evaluate the potential heavy metal contamination of long-used (500+ years) church necrosol graveyards with different soil type, the graveyards datasets acquired from surface soil, shallow soil depths (>0.75m) and from the empty grave (>2m) (at study site 1 only) all showed heavy metal concentrations that were far higher than background control samples taken ~100m away from both graveyards (Tables 1-5), with measured values well above the threshold level (75 ppm) identified for potential ecotoxicological effects in soils identified by deVries et al. (2007). Indeed in many of the graveyard soil sampling points examined, lead concentrations were also above the predicted 'no effect' concentrations of 166 ppm and 212 ppm reported by Smolders et al. (2009) and the European Chemicals Agency (ECA - Ecotoxicological Summary for lead), respectively. This paper showed heavy metals were present in the two study site soils at much higher concentrations than those found in other studies, with Neckel et al. (2016), for example, only recording lead values up to 127 mg/kg (Table 6). Fiedler et al. (2012) graveyard study evidenced comparable lead levels from this study to an uncovered coffin (Table 6). It should be noted that arsenic concentrations were generally low at both study sites and did not exceed either control values or WHO soil standards of ~10 mg/kg, even though it is not very mobile in soil (Fiedler et al. 2012).

There were also large variations of measured heavy metal element concentrations within each graveyard, with the highest values being found generally in soil samples taken adjacent to church buildings. Concentrations decreased, with increasing spatial distance from the churches. This trend in lead distributions could be partially explained by soil adjacent to church buildings containing relict lead flashings and roof linings, but other elements also showed this trend (e.g. calcium), which may suggest a higher burial density nearest the church and corresponding release of elements from coffins as Fiedler et al. (2012) measured from graveyard coffins in Germany (Table 6). Generally, soil samples showed the same trend of decreasing element concentrations with increasing soil depth, except for lead at study site 1 with sandy soil which had consistently high lead values down to 0.75m.

Comparing the two graveyard study sites, although the number of known burials and graveyard areal extent was different, the actual burial density was similar (~2.5 m²), so element concentration differences may be due to the different soil type. Surface soil (and down to 0.25m bgl) heavy metal concentrations were generally higher in the clay soil at study site 2, when compared to the sandy soil at study site 1, suggesting elements are less mobile in the low porosity/permeability clay soils. In contrast, higher element concentration values occur in deeper soils in the sandy study site 1 but unfortunately deeper samples than 0.75m bgl were not collected from the clay soil at study site 2 so it cannot be stated definitively that higher element levels are not present at depth in this graveyard.

This study has important implications for managing both historical and contemporary burial grounds, in relation to re-use and potential environmental and ecological contamination impacts from burial sites. Depending upon the soil type, as evidenced here, mobile heavy metals may leach away from the graveyard area itself and potentially to nearby surface and groundwater supplies if the geological conditions are suitable for this. Finally, a number of closed churches, graveyards and cemeteries are being deconsecrated and turned into residential dwellings, and soil analysis for heavy metal concentrations would be highly recommended especially if people living in these areas wanted to grow edible produce which could bioaccumulate these heavy metals in their tissues. Further research is needed on these converted site to assess this important environmental and human health contamination risk.

Future work

Whilst one empty grave at the study site 1 was able to be sampled, it would obviously be advantageous to sample grave soil deeper and ideally adjacent to coffins themselves as per Fiedler et al. (2012) adipocere study. Other burial ground types, for example green or 'natural' burials, are becoming increasingly popular globally, with 270 UK sites being built between 1993-2015 alone (Yarwood et al. 2015). These generally have lower burial densities, when compared to cemetery/graveyard burial grounds, biodegradable receptacles (e.g. shrouds, cardboard or wicker-based), but involve more shallow or even vertical burials (Kim et al., 2008). These factors would suggest early decomposition stages releasing more fluids, including embalming fluids, into the surrounding soil, when compared to more traditional burials, with accompanying increased surrounding soil contamination, but little research has been undertaken on this to-date.

Conclusions

Portable pXRF *in situ* surface surveys can collect large quantities of data with minimal sample preparation compared to other analytical analyses techniques, which may range from none (*in-situ*, non-destructive analysis) to extensive (sample collection, homogenisation, and pelleting). In addition, rapid *in situ* surface analysis techniques can identify priority areas for subsequent follow-up urgent inspection if concentrations for potential ecotoxicological effects are above threshold levels in soils.

This paper provides two case studies of long-used (500+ years) burial grounds, UK church graveyards in this case, whose necrosols are contaminated by heavy metals. In particular, lead was found in concentrations well in excess of current environmental guidelines, although these concentrations are not uniformly distributed, both in extent across the graveyards and in depth below ground level. The highest levels of contamination are in the top 0.25 m and adjacent to church structures, potentially due to high burial concentrations and/or due to relict church materials. This will be important for burial ground management, those living adjacent to burial grounds, potential surface/groundwater contamination and where burial grounds have been deconsecrated and turned into residential dwellings.

This paper is limited by only studying two UK graveyards, albeit long-used with different soil textures, and by the numbers of soil samples collected, analysed and measured. However, the implications for other church graveyards to be similarly contaminated is clear. More accurate analytical equipment should be used to refine these initial results and obtain absolute element measurements. Further research should investigate other graveyards, cemeteries, green burials and other burial grounds with different burial ages, in other soil types, as well as collecting soil within and adjacent to graves.

Declarations

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Competing interests

The authors declare that they have no competing interests

Authors contributions

C.M. undertook the desk study, collected field and laboratory data and performed the literature review, with M.E. collecting field/lab data and analysis. J.P. designed the study, wrote the initial draft and co-ordinated authors. A.J. assisted with data analysis, calibrations and co-wrote the paper. K.W. assisted with field data collection, figure generation and co-wrote the paper. V.J., I.O., H.G., IGS and J.G. co-wrote the paper.

Ethics approval and consent to participate

This project has passed the Keele University's research ethics panel review.

Consent to publish

Not applicable

Availability of data and materials

pXRF data and burial records for the two respective case study sites are available on Keele's eRepository, the DOI link of which is: <http://doi.org/10.21252/sjkk-w810>

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Supplemental Data

The Supplemental Data is not available with this version

Figures



Figure 1

Schematic drawing of burials types showing, a) single isolated earth-cut grave with dominantly wooden (rarely metal) coffin, b) multiple vertically-stacked earth-cut graves, c) isolated brick-lined grave with coffin, d) multiple stacked brick lined grave, e) grave vault with multiple stacked brick lined burials and, f) 'green' burial (modified from Hansen et al., 2014)



Figure 2

Study site 1 map at St. John's Church sandy soil graveyard (red box), Keele, Staffordshire, with UK location (inset)



Figure 3

Study site 2 map at St. Michael and All Angels' Church clay-rich graveyard (red box), Stockton, Norfolk, with UK location (inset)

Figure 4

Schematic diagram showing the generalized soil hand-auger profile results at (a) study site 1 at St. John's Church sandy soil graveyard, Keele, Staffordshire, UK, and (b) study site 2 at St. Michael's Church clay-rich soil graveyard, Stockton, Norfolk, UK

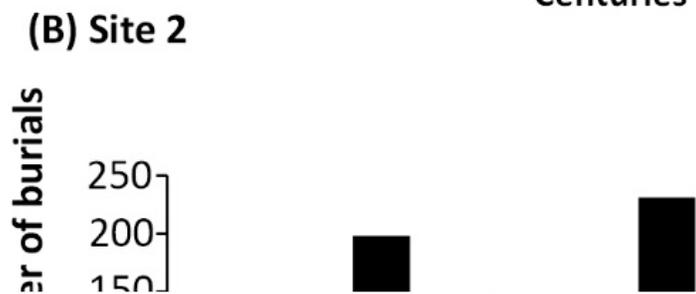
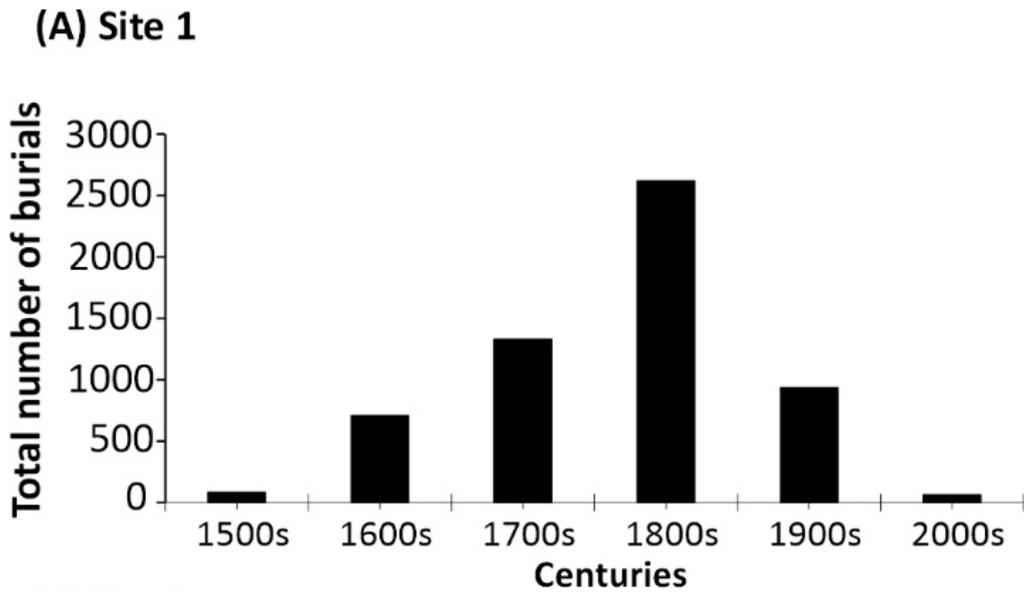


Figure 5

Summary plot showing known burial records of (a) 5,735 burials (1585-2018 CE) at study site 1 St. John's Church, Keele, Staffordshire, UK, and (b) 669 burials (1561-2018 CE) at study site 2 St. Michael and All Angels' Church, Stockton, Norfolk, UK

Figure 6

(A) selected sample location with overlying vegetation/pebbles cleared and sample bag shown. (B) Field pXRF 120 s analysis of a sampled surface graveyard soil location. (C) 0.75 m soil auger used 3x at each location to collect 0-25cm, 26-50cm and 51-75cm depth ranges soil samples. (D) pre-dug grave that was able to be sampled every 0.25 m down to 2 m bgl. (E) 20 Tn soil press used to produce (F) soil pellet, see text for details

Figure 7

Study site 1. a) In-situ surface and b) laboratory soil pellet lead element pXRF concentrations. Dashed line marks graveyard extent

Figure 8

Study site 2. a) In-situ surface and b) laboratory soil pellet lead element pXRF concentrations. Dashed line marks graveyard extent

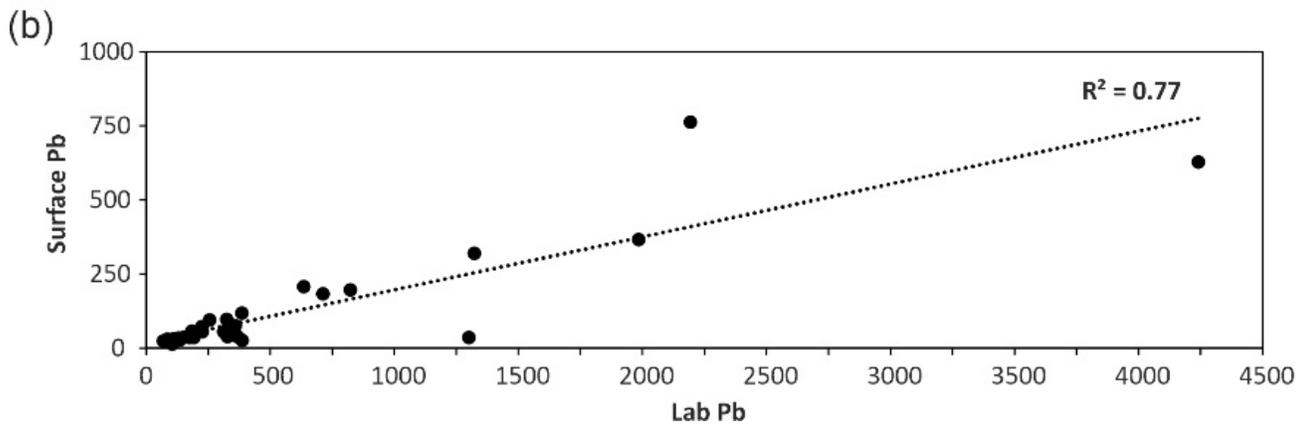
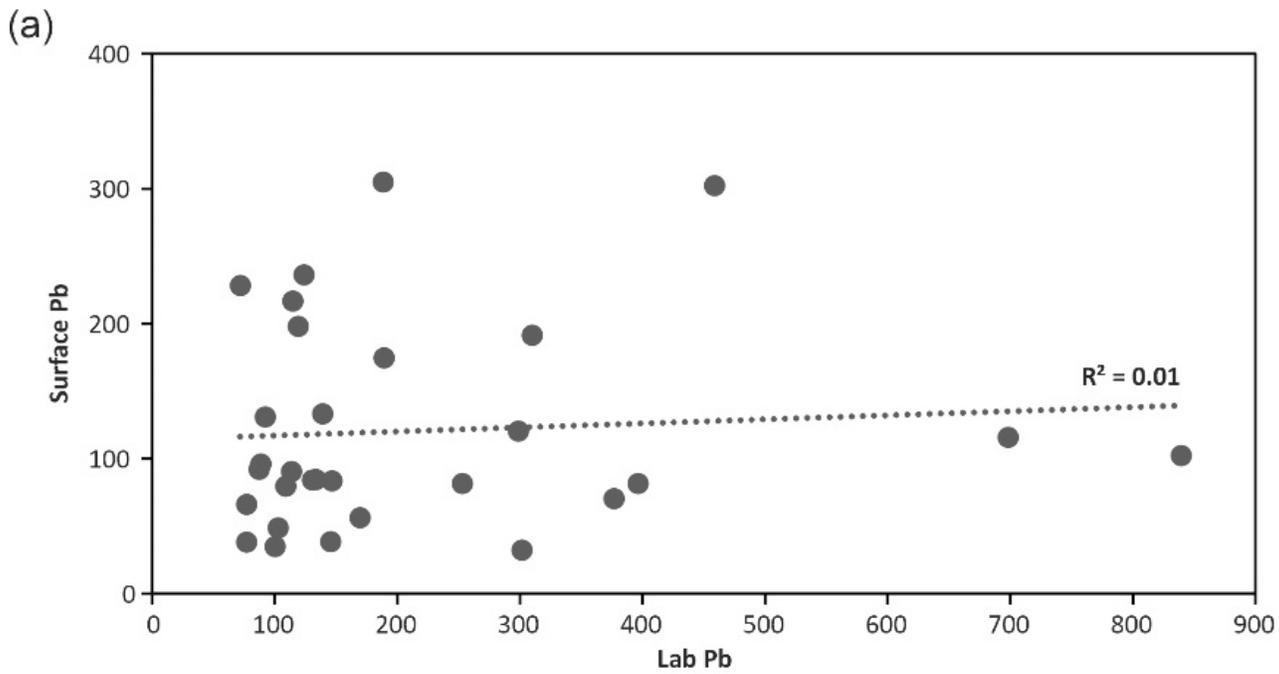


Figure 9

Graph cross-plots of (a) in-situ soil and (b) laboratory soil pelletised measured lead concentrations for (a) study site 1 sandy soil and (b) study site 2 clay soil respectively

Figure 10

Graphs showing lead and calcium element pXRF surface laboratory soil pellet pXRF concentrations with distance from a) study site 1 and b) study site 2. Note logarithmic trendline with R2 fit is shown

a) Site 1 sandy soil

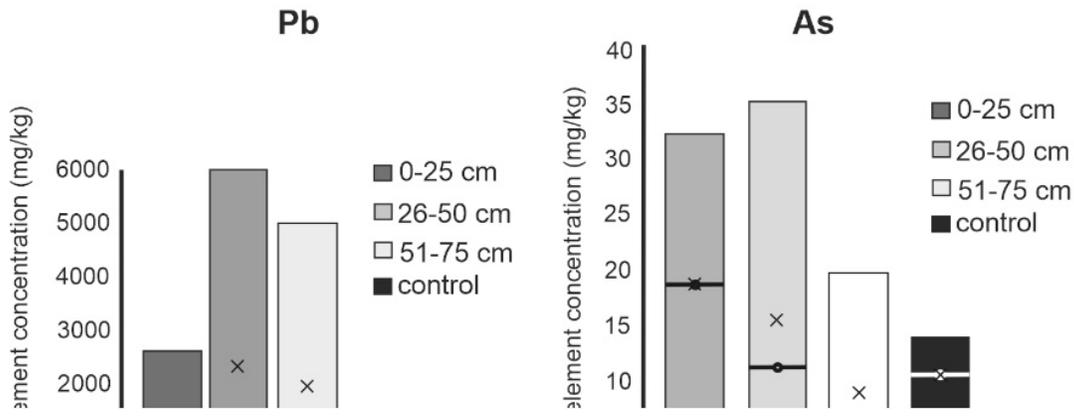


Figure 11

Box-whisker graph plots of lead and arsenic element pXRF concentrations from soil auger pellets from the different depths investigated (see keys) from a) study site 1 sandy soil, and b) study site 2 clay soil graveyards

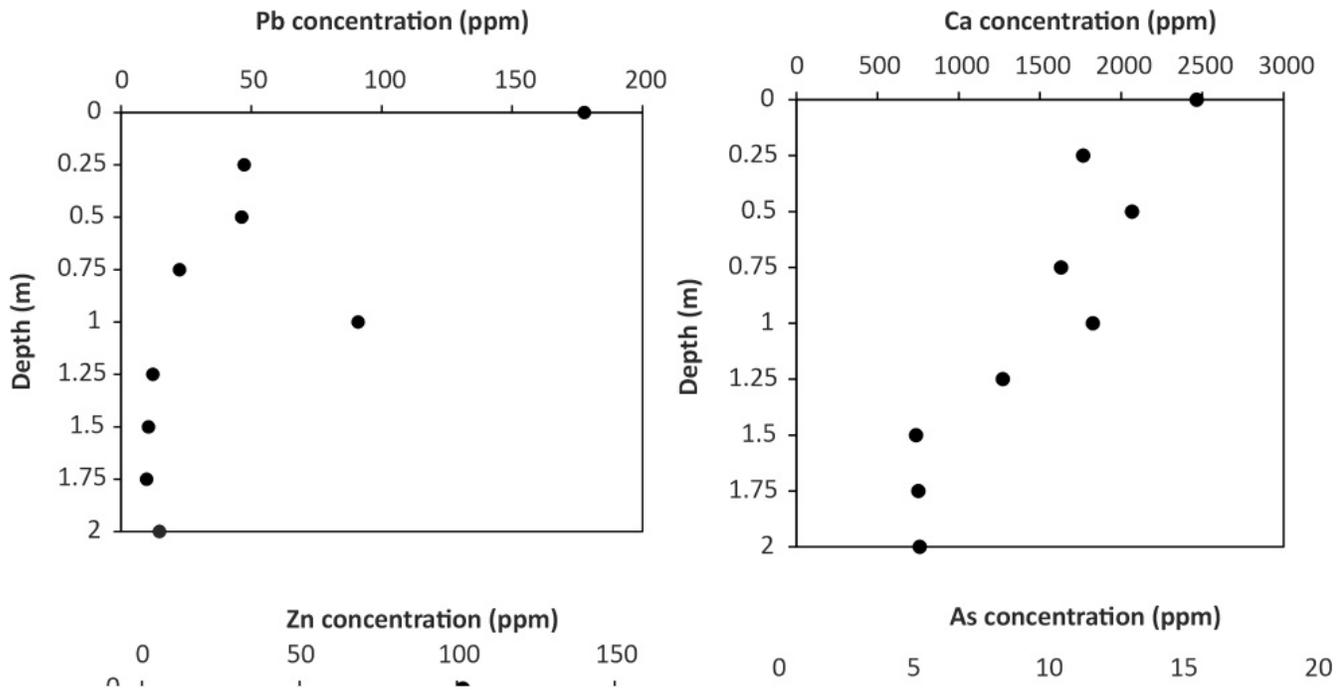


Figure 12

Graphs of selected element pXRF concentrations against empty grave depths bgl at study site 1 St. Johns graveyard sandy soil graveyard, Keele, Staffordshire, UK