Development of a searchable major and trace element database for use in forensic soil comparisons

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ABSTRACT

Forensic soil comparisons are normally undertaken on the basis of several physical, chemical and biological properties, but in all cases the interpretation of results is dependent on the availability of relevant contextual information. This paper summarises the results of major and trace element analyses performed using inductively coupled plasma atomic emission spectrometry and inductively coupled plasma-mass spectrometry on the <150 µm size fraction of 1896 soil samples collected in connection with forensic investigations in England and Wales between 1999 and 2007. A number of new methods are described which facilitate inter-sample comparison. Although the available data do not provide uniform geographical coverage they do provide useful information which can assist police search investigations and they provide valuable contextual information which aids the evidential assessment of soil evidence when used in court. There is considerable scope to improve the database by increasing the geographical coverage and increasing the number of soil attributes which are included.

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1. Introduction

The forensic investigation of soil and other forms of physical trace evidence involves two stages: (1) comparison of a questioned sample with one or more samples of known origin, and (2) evaluation of the significance of observed similarities and differences in order to arrive at a conclusion regarding a possible association. The latter depends heavily on the availability of background information relating to the frequency of occurrence and geographical distribution of samples which show similar levels of similarity or difference to those being compared. Comparison of soils and sediment samples normally involves consideration of several different properties, including particle size distribution, colour, particle typology, mineralogy, chemical composition, pollen assemblages and other microfossils [1–3]. Many techniques have been developed over the past 20 years which allow these properties to be quantified, in many cases with good accuracy and precision. However, there are currently no comprehensive databases to assist interpretation of the data obtained. Useful contextual information at the national and regional scale is available in the form of maps, atlases and descriptive compendia of soil properties, but the supporting data are often presented in a form which limits their

use in forensic investigations. For example, a number of soil and sediment sampling programmes have been carried out in England and Wales over the past 40 years, principally for purposes of geochemical mapping, mineral prospecting and land-use assessment purposes [4–10], and similar exercises have been undertaken in continental Europe [11], but the data mostly refer to composite samples representing relatively large areas (1 km^2 or more). To date there have been few published attempts to develop databases of soil and sediment properties specifically with forensic applications in mind (e.g. [12]).

All databases relating to physical evidence are subject to limitations imposed by the number of samples they contain, the degree to which those samples are representative of the populations from which they are drawn, the type and quality of the information included, the discriminatory capacity of the criteria which can be applied to the data, and the fact that some types of information may be rendered out of date very quickly. Nonetheless, databases can provide useful sources of contextual information which aids interpretation of potential evidential significance [13].

This paper summarises an ongoing attempt to develop a database containing information about the major and trace element compositions of surface and near-surface soil samples in England and Wales, and describes a number of methods which have been developed to compare data for questioned samples with new control samples and existing samples in the database. Information about soil properties other than chemical composition is also recorded, and in casework investigations comparisons are almost always made on the basis of several criteria. For purposes of illustration in the present paper, reference is made only to major and trace element compositional

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data acquired from the <150 μm size fraction of 1896 soil samples collected from known locations in England and Wales between 1999 and 2007. Results for samples collected since that time, or which relate to different size fractions, locations outside England and Wales, and to soil recovered from suspect vehicles, footwear, implements etc., are not considered here.

2. Materials and methods

The samples referred to in this paper were collected from numerous different locations in England, and some in Wales (Fig. 1). The sampling locations are not uniformly distributed and were not selected according to a pre-defined sampling framework; rather, they reflect locations of interest in casework investigations during the period 1999–2007. In some of the areas shown, relatively large numbers of samples (>50) were collected within an area of a few square kilometres, while in other areas the number of collected samples was much smaller, or samples were collected across a wider area (10–100 km²). As such, the dataset is not considered to be statistically representative of the land area of England and Wales as whole. Rather, the data should be considered in a similar way to the 'survey' information often used in connection with other forms of physical trace evidence, such as the frequencies with which particular fibre types have been recorded during laboratory examination of submitted clothing items.

The great majority of the samples analysed were taken from the ground surface (0–5 cm depth), although approximately 3% were collected from shallow pits and graves (<50 cm depth). Most were collected using a stainless steel trowel or similar instrument which was thoroughly cleaned with distilled deionised water before and after each sample was taken. In the laboratory, each

sample was homogenized by mixing prior to sub-sampling for a range of analysis types. The <150 μm size fraction was separated from one of the sub-samples in each case by wet-sieving though disposable nylon mesh using distilled de-ionized water [14,15]. The <150 μm size fraction was selected partly because it has been used extensively in previous geochemical investigations and partly because tests have demonstrated that this size fraction is practically efficient to obtain, normally yields sufficient material for analysis from small forensic samples, and frequently provides a good 'proxy' for the bulk elemental composition of sediments and soils [16].

The concentrations of up to 10 major and 49 trace elements were determined using inductively coupled plasma atomic emission spectrometry (ICP-AES) and inductively coupled plasma-mass spectrometry (ICP-MS) at two laboratories: the Department of Geology, Royal Holloway, University of London (RHUL), and the School of Earth Sciences, University of Greenwich (UoG). Some samples were analysed at both laboratories, although most were analysed at only one. Approximately a third of the samples were analyzed only by ICP-AES, but the majority were analyzed using both ICP-AES and ICP-MS to provide data for a wider range of elements. The RHUL data were generated using a Perkin-Elmer Optima 3300 RL ICP-AES instrument and a Perkin-Elmer Sciex Elan quadropole ICP-MS instrument. The UoG data were generated using a TGA Iris Advantage instrument (ICP-AES) and a Thermo-Elemental X7 instrument with an HPI interface (ICP-MS). The sample preparation, data processing and quality control procedures used in the two laboratories, which include regular analysis of international standard materials and laboratory reference materials, have been described in previous publications [15,17,18]. Owing to improvements in analytical methods over the period of study, the number of trace elements determined has increased over time. As a result, the number of samples for which elemental data are available ranges from 337 (thulium) to 1896 (all of the major elements except silicon). The concentration s of major elements are reported in terms of weight percent oxide (wt.%) and the concentrations of trace elements in μg g^{-1}.

The compositional data were collated in Microsoft® Excel and analysed using Microsoft XLSTAT®. The data generated by each laboratory can be interrogated separately or in combination, using major elements only, trace elements only, elemental ratios only or various combinations of these. For the purposes this paper, only a limited number of comparative examples are presented, using data for both laboratories and a combination of major and trace element concentrations and ratios.

3. Results

3.1. Inter-laboratory comparison

Table 1 summarises the results for 10 samples which were analysed at both laboratories (different sub-samples of the same ground powder, prepared as separate solutions at each laboratory). The results show relatively small differences between the two laboratories in terms of the mean concentrations of the ten major elements and most of the trace elements. Only three elements (Ta, Pb and Co) showed relative differences in the mean concentrations recorded by the laboratories larger than 20%, and eight others (Ti, P, Cs, Hf, U, Th, Ho, Er) showed relative differences larger than 10%. The inter-laboratory differences in major oxide concentrations and elemental ratios derived from the primary data were also small, with only the U/Th ratio showing a relative difference larger than 10% (Table 1). This level of variation is no greater than that found during analysis of different sub-samples taken from the same parent sample at a single laboratory over time [15]. It is therefore considered justified to combine the
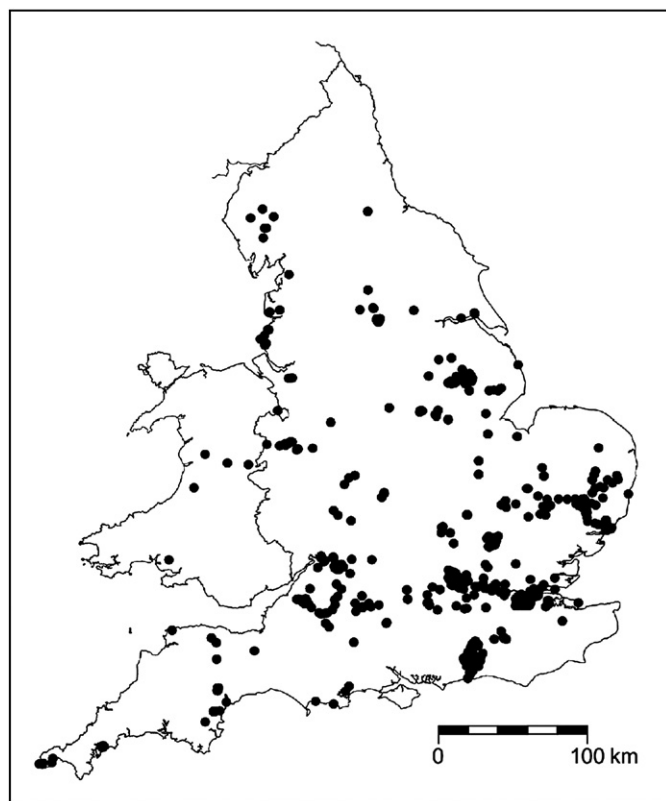

Fig. 1. Geographical distribution of the 1896 soil samples from England and Wales contained in the dataset. Note that the locations shown include groups of several samples.