

1 **Automated mineralogical profiling of soils as an indicator of local**  
2 **bedrock lithology: a tool for predictive forensic geolocation**

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4 Duncan Pirrie<sup>a,d</sup>, Daniel E. Crean<sup>b</sup>, Allan J. Pidduck<sup>b</sup>, Timothy M. Nicholls<sup>b</sup>,  
5 Roy P. Awbery<sup>b</sup>, Robin K. Shail<sup>c</sup>

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8 <sup>a</sup>Helford Geoscience LLP, Trelowarren Mill Barn, Mawgan, Helston, Cornwall,  
9 TR12 6AE, UK

10

11 <sup>b</sup>AWE Plc, Aldermaston, Reading, RG7 4PR, UK

12

13 <sup>c</sup>Camborne School of Mines, University of Exeter, Penryn Campus, Penryn,  
14 Cornwall, TR10 9FE, UK

15

16 <sup>d</sup>School of Applied Sciences, Faculty of Computing, Engineering and  
17 Sciences, University of South Wales, Glyntaf, Pontypridd, Rhondda Cynon  
18 Taff, CF37 4AT, UK

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20 **ABSTRACT**

21

22 The use of soil evidence to identify an unknown location is a powerful tool to  
23 determine the provenance of an item in an investigation. We are particularly  
24 interested in the use of these indicators in nuclear forensic cases, whereby  
25 identification of locations associated with for example, a smuggled nuclear  
26 material, may be used to indicate the provenance of a find. The use of soil  
27 evidence to identify an unknown location relies on understanding and  
28 predicting how soils vary in composition depending on their geological /  
29 geographical setting. In this study, compositional links between the  
30 mineralogy of forty soils and the underlying bedrock geology were  
31 established. The soil samples were collected from locations with broadly  
32 similar climate and land use across a range of geological settings in a 'test  
33 bed' 3500 km<sup>2</sup> area of South West England. In this region, the soils formed  
34 through chemical weathering of the bedrock, representing a worst case for

35 this type of forensic geolocation due to the high degree of alteration of the  
36 parent rock during soil formation. The mineralogy was quantified using  
37 automated SEM-EDX analysis. The soil mineralogy and texture are consistent  
38 with the underlying geology as indicated by regional-scale geological  
39 mapping.

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41 Keywords: soil forensics, provenancing.

42

### 43 **Introduction**

44

45 It is widely recognised that soil is a useful class of trace evidence which  
46 can be used to test a possible association between a geographical location  
47 and material recovered from, for example, footwear, clothing, vehicles and  
48 objects used during an offence (e.g. Bull et al., 2006; Pirrie and Ruffell, 2012).  
49 In general, most published forensic soil science studies have addressed the  
50 use of soil as an evidential tool, testing an association between an object and  
51 a known location through comparative analysis (Fitzpatrick et al., 2009).  
52 However, soil analysis can also be used to attempt to identify unknown  
53 locations based on the soil characteristics (Bowen and Craven, 2013; Lark  
54 and Rawlins, 2008; Owens et al., 2016). This use of soil trace evidence to  
55 determine the geographic position of otherwise unknown locations can be  
56 referred to as predictive geolocation (Pirrie et al., 2017) and is of increasing  
57 interest to the forensic geoscience community (e.g. Stern et al., 2019; Caritat  
58 et al., 2019).

59 One aspect of our interest in predictive geolocation relates to its  
60 potential to determine the geographic source (provenance) of an item in a  
61 nuclear forensic investigation (Mayer et al., 2012). These scenarios involve  
62 the discovery or interdiction of nuclear material outside regulatory or  
63 institutional control, such as so-called 'nuclear smuggling' cases (Wallenius et  
64 al., 2006; Mayer et al., 2015). In these investigations, there may be a wide  
65 range of potential sources of the material, (e.g. facilities which have  
66 processed the particular type of material), and therefore the potential to  
67 constrain the possible origins of a find through predictive geolocation analysis  
68 would be of great value in establishing the provenance of a found material.

69 This is complementary to the types of analysis which fall under the field of  
70 nuclear forensic science (Keegan et al., 2016), which relate to the properties  
71 of the material itself, such as the identification of particular processes through  
72 the evaluation of signature chemical properties of the material.

73 Predictive geolocation analysis relies on signatures accumulated from  
74 the environments to which a nuclear material has been exposed during  
75 production, transport and storage; both before and after institutional control  
76 was lost. Packing and repackaging of materials may introduce a number of  
77 internal interfaces where materials from different environments may  
78 accumulate. These environmental signatures may include natural materials  
79 such as soils and dusts, to which forensic soil geolocation analysis techniques  
80 can be applied to indicate location(s) associated with the sample.

81 For this predictive geolocation analysis to be practical, it is necessary  
82 to select key parameters which can be measured in a realistic (short)  
83 timescale for use in an investigation, and which have a high degree of  
84 specificity in indicating an unknown location. Owing to the wide availability of  
85 spatial geological reference datasets (e.g. from national geological surveys),  
86 indicators related to the underlying lithology at a location are useful in this  
87 context (Pirrie et al., 2013). Determination of the likely source geology from  
88 soil analysis would allow an investigation to be focussed to areas where this  
89 setting is known to occur. For this approach to have forensic validity, the  
90 relationship between soil mineralogy and underlying geology has to be  
91 established. Soil-forming processes can profoundly alter the composition of  
92 the parent material depending on environmental conditions, and therefore  
93 there is a need to understand the specificity with which lithological signatures  
94 could be detected in a range of soil forming environments.

95 In this paper we present a systematic study of the relationship between  
96 the underlying geology and soil mineralogy in a geologically varied 3500 km<sup>2</sup>  
97 area of SW England. The area provides a 'test bed' with a variety of well  
98 documented major rock types and settings. Soils within the area are mainly  
99 mature loams, developed through chemical weathering and human  
100 management. This represents an advanced state of alteration of the  
101 underlying rock and therefore a high potential for disturbance of the  
102 mineralogical signatures linking the soil to the associated bedrock. Automated

103 mineralogy based on scanning electron microscopy was used as a rapid  
104 method for characterisation the mineralogy and texture of the soils. This tests  
105 the link between rapidly measureable soil characteristics (in this case  
106 mineralogy) and widely available geological datasets, such that an indication  
107 of potential geological settings could be provided from an unknown soil  
108 sample in an investigation.

109

## 110 **Overview of Study Area**

111

112 In this study 35 geographical locations over an area of approximately  
113 3500 km<sup>2</sup> in Cornwall and Devon, SW England, UK were selected for soil  
114 sampling (Fig. 1, Table 1). Factors such as variations in climate and land-use  
115 were minimised as far as possible, such that the dataset allows the  
116 significance of the underlying bedrock geology as the main control on soil  
117 mineralogy to be tested. All of the sampling locations would also have had  
118 surface superficial sediments present mantling the bedrock geology to  
119 variable extents.

120 The selected sampling area whilst geologically varied, shows the  
121 repetition of a number of major rock types, and samples from these were  
122 taken to test discrimination between soils developed on different age units  
123 composed of the same dominant rock types. SW England has also been a  
124 recent focus for baseline environmental and geochemical surveys based on  
125 both airborne datasets and regional surface soil and sediment sampling  
126 programmes (Beamish, 2015; Kirkwood et al., 2016).

127

## 128 *Geology*

129

130 SW England is an Upper Palaeozoic massif whose bedrock geology, in  
131 common with much of western Europe, was strongly influenced by the late  
132 Palaeozoic Variscan mountain-building episode (Shail and Leveridge, 2009).  
133 The near-surface geology is dominated by Devonian and Carboniferous  
134 metasedimentary rocks (Fig 1.). Other significant features include associated  
135 minor basic intrusive / extrusive igneous rocks, a major granite batholith which  
136 was emplaced in the Early Permian, and the Lizard Complex, which includes

137 partially serpentinised mantle peridotites, gabbros and high-grade  
138 metamorphosed mafic igneous rocks. Devonian and Carboniferous  
139 successions occur in six east-west trending sedimentary basins (Leveridge,  
140 2011; Leveridge and Shail, 2011a, b). These formed during rifting which  
141 resulted in the formation of a passive margin and associated oceanic  
142 lithosphere. Sedimentation in the Gramscatho and Culm basins continued  
143 during the early stages of the Variscan continental collision (Leveridge, 2011;  
144 Leveridge and Shail, 2011a). The Looe Basin includes non-marine  
145 sandstones and mudstones (Leveridge, 2011) but, more generally, the basin  
146 successions comprise mudstones and sandstones, along with minor  
147 limestones and cherts, deposited in shallow to deep-marine environments  
148 (Leveridge and Shail, 2011b). Rift-related mafic igneous rocks (basalts,  
149 dolerites and gabbros) are locally important constituents of the basin fills and  
150 sedimentary exhalative (SedEx) and volcanic massive sulphide (VMS)  
151 mineralisation styles locally occur (Benham et al., 2005). The Lizard Complex  
152 includes partially serpentinised mantle peridotites, gabbros and high-grade  
153 metamorphosed mafic igneous rocks; it represents a fragment of the oceanic  
154 lithosphere (an ophiolite) formed during rifting and also includes small areas of  
155 high-grade continental metamorphic rocks.

156 Variscan continental collision brought about deformation and low-grade  
157 regional metamorphism of all of the basinal successions (Shail and Leveridge,  
158 2009). Consequently the rocks are variably thrust-faulted and folded and one  
159 or more cleavages are developed. Quartz veins, precipitated from  
160 metamorphic fluids, are ubiquitous and locally associated with precious metal  
161 mineralisation. Continental collision continued until the latest Carboniferous,  
162 when it was followed by a NNW-SSE extensional regime which persisted  
163 through most of the Early Permian. This extension led to the development of  
164 post-Variscan Permian 'red-bed' sedimentary basins (Edwards et al., 1997)  
165 and voluminous post-collisional felsic magmatism. The principal expression of  
166 the latter is the Cornubian Batholith which comprises a variety of granite types  
167 and is associated with rhyolite / microgranite dykes known locally as 'elvans'  
168 (Simons et al., 2016). Host rocks within 1 km or so of the batholith margins  
169 exhibit contact metamorphism. The granites, elvans and their surrounding  
170 host rocks are cut by extensional fault systems hosting Early-Mid-Permian

171 polymetallic W-Sn-Cu-As-Pb-Zn magmatic-hydrothermal mineralisation, the  
172 working of which has resulted in substantial areas of metal-contaminated  
173 made ground throughout the region (Pirrie et al., 2003). A subordinate Mid-  
174 Triassic fault-controlled epithermal mineralisation episode, associated with the  
175 migration of basinal brines from 'red-bed' basins, resulted in localized Pb-Zn-  
176 barite mineralization (Simons et al., 2011).

177 The Quaternary geological history of SW England is dominated by  
178 repeated intervals of periglacial climates and warmer inter-glacials. Although  
179 some authors have speculated on the possibility of small cirque glaciers  
180 (Harrison et al., 1998), there is no evidence for significant glaciation  
181 throughout the region during the repeated cold climate intervals of the  
182 Quaternary. Instead the bedrock geology was influenced by periglacial  
183 processes with down-slope mass wasting of the local bedrock units. These  
184 locally derived periglacial sediments are referred to as "head deposits" and  
185 typically thicken into valleys. Along the north coast marine-derived carbonate  
186 blown sand deposits are locally developed as dune sand systems. Flooded  
187 steep sided valley systems known as rias, now forming major estuaries, are  
188 developed along the south coast (e.g. the Helford, Fal, Fowey and Tamar  
189 estuaries) along with smaller scale systems on the north coast (Hayle, Gannel  
190 and Camel estuaries). These estuaries have received significant volumes of  
191 contaminated mine waste as a result of the historical mining activity in the  
192 region (Pirrie and Shail, 2018).

193 For the purposes of presentation and analysis of results, six broad  
194 lithological groupings were identified from geological maps into which the  
195 sampling locations were assigned. These were granitic (samples 1, 3, 10, 17,  
196 20 and 34), mafic-igneous (samples 2, 12, 15, 22, 32, 35), metamorphic  
197 (samples 4, 5, 14, 16), metamorphosed mudstone (samples 6, 7, 8, 13, 18,  
198 19, 21, 23, 24, 26, 27, 28, 29), metamorphosed sandstone (samples 9, 11, 25)  
199 and Carboniferous sedimentary (samples 30, 31 and 33).

200

### 201 *Geography, climate and soils*

202

203 Despite the historical mining industry throughout the region, Cornwall  
204 and Devon are today predominantly rural environments dominated by

205 agricultural land use. Lower lying areas are typically underlain by the  
206 Devonian and Carboniferous metasedimentary rocks whilst the granites  
207 underlay upland moorland which has an elevation of up to 621 m on  
208 Dartmoor. The region has an oceanic climate, with a small range in annual  
209 mean temperature between approximately 6°C (winter) and 16°C (summer),  
210 mild winter temperatures and among the highest wind speeds in the United  
211 Kingdom. Winters are wet and summers dry with annual precipitation totals of  
212 900–1,000 mm at the coast, although annual precipitation increases inland, to  
213 up to 2,000 mm on the granite moorland (Kosanic et al., 2014).

214 According to the available soil classification schemes, the soil types  
215 present show relatively little variation within the studied area  
216 ([www.landis.org.uk/soilscapes](http://www.landis.org.uk/soilscapes)). The majority of the Devonian and  
217 Carboniferous metasedimentary rocks are overlain by acidic loamy soils,  
218 slightly acidic loamy soils or slightly acidic base-rich soils. The granites are  
219 overlain by the same acid loamy soils, along with very acid loamy upland soils  
220 with a wet peaty surface, very acid upland soils with a peaty surface, and on  
221 Bodmin Moor and Dartmoor blanket bog peat soils. Extensive areas on the St  
222 Austell Granite are restored soils as a result of the china clay (kaolinite)  
223 mining in this area. The soils on the Lizard Complex comprise slightly acidic  
224 base-rich soils and wet acid loamy and clayey soils.

225

## 226 **Materials and methods**

227

### 228 *Soil sampling*

229

230 Prior to sampling, 35 rural locations throughout Devon and Cornwall  
231 with (a) different underlying bedrock geology but (b) similar land-use, were  
232 identified as suitable for sampling based on examination of topographic maps,  
233 British Geological Survey geological maps and Google Earth imagery. Soil  
234 sampling was carried out between the 24th February and the 7th March 2016.  
235 Wherever possible, areas of managed grassland, ploughed and re-seeded for  
236 grazing, were selected for sampling. Sampling sites within these areas were  
237 selected away from sources of apparent introduced geological materials such  
238 as tracks and roadways. Localised areas of historic mining activity are

239 widespread in SW England; however, these were also deliberately avoided as  
240 mineral contamination introduced into the surface environment near these  
241 sites is highly distinctive, and therefore not representative of the underlying  
242 bedrock-soil relationship (Pirrie et al., 2003). In the context of geolocation,  
243 identification of these distinctive mineral species would allow an unknown  
244 area to be identified with high precision; however, these signatures do not  
245 have wider relevance in localities without a significant history of mining  
246 contamination.

247 At each sampling location a 250 m long, linear transect was  
248 established. In some cases, transects were offset to take into account the  
249 orientation of field boundaries. Five samples were collected in total along the  
250 line of the transect, with the first sample at the start (0 m), followed by  
251 samples taken at distances of 10 m, 50 m, 100 m and 250 m along the  
252 transect. Sampling location co-ordinates were recorded using a handheld  
253 GPS.

254 To extract the soil sample, a labelled 35 mm diameter clean plastic  
255 sampling pot was pushed approximately 2 cm into the ground surface (Fig. 2).  
256 The edges of the pots are thin and effectively cut a clean edge into the soil  
257 profile. If possible, areas of exposed soil were preferentially selected for  
258 sampling. The *in-situ* sampling pots were digitally photographed, removed  
259 and then sealed with tamper evident lids and placed within zip lock plastic  
260 bags. The pots were then opened so that the exposed visible surface  
261 represents the lowest part of the sampled soil, estimated to be at a depth of  
262 between 1 and 2 cm below the surface. A subsample was removed from this  
263 lower surface and dried for 2 hours at a temperature of 50°C, before being  
264 sealed within a zip-lock plastic bag. In total, 175 soil samples were collected,  
265 subsampled and dried.

266

### 267 *Mineral analysis*

268

269 The initial (0 m) sample from each of the 35 locations was selected for  
270 mineral analysis (Table 2). In addition, one sampling location was selected at  
271 random (location 24, Porthtowan Formation metamorphosed mudstone) and  
272 all 5 samples collected from that location were also prepared for analysis.

273 Each soil subsample was gently disaggregated, placed into a 30 mm mould  
274 and mixed with epofix resin. The samples were allowed to cure for 24 hrs,  
275 labelled and back-filled with araldite resin and heated at 50°C for 2 hours.  
276 The blocks were then polished and carbon coated prior to mineral analysis.  
277 Mineral analysis of the 40 samples was carried out using automated scanning  
278 electron microscopy (SEM) with linked energy dispersive X-ray (EDX)  
279 spectrometers, based on QEMSCAN technology.

280 Automated SEM-EDS is a widely used method for mineral analysis  
281 (Armitage et al., 2010; Williamson et al., 2013; Eby et al., 2015) and has  
282 previously been used for the forensic analysis of soil samples (Pirrie and  
283 Rollinson, 2011; Pirrie et al., 2004, 2013, 2014). In brief, individual particles  
284 are located on the cut face of the polished block and are then phase mapped  
285 in cross section by the acquisition of energy dispersive X-ray spectra (in this  
286 study using 1000 counts/pixel) at a regular, operator defined spacing across  
287 the sample. The 1000 count spectra places limits of detection for elements  
288 present within the individual mineral grain at an abundance of approximately 3  
289 atom percent (Andersen et al., 2009). In this study spectra were acquired  
290 across the sample using a 6 µm spacing, and the particle size range accepted  
291 for analysis was 9 µm to 800 µm. Each measured spectrum is assigned to a  
292 mineral name or chemical grouping by matching the elemental signature  
293 against a user-defined classification scheme (Pirrie and Rollinson, 2011). The  
294 mineral categories used in this study are provided in Table 3.

295 In each soil sample, >5000 individual mineral grains were analysed,  
296 based on the acquisition of between 50,734 and 487,685 EDS analysis points  
297 per sample. The number of EDS analyses is controlled by the size of the  
298 individual soil particles, such that finer-grained particles are effectively  
299 mapped by a smaller number of spectra than a larger particle (Table 2). Each  
300 sample took between 20 and 60 minutes to be analysed, showing that the  
301 methodology is sufficient for rapid sample screening. The automated  
302 mineralogy data were acquired using an FEI 650F scanning electron  
303 microscope, with an accelerating voltage of 25kv, equipped with twin Bruker  
304 30 mm<sup>2</sup> Quantax spectrometers, and the data were processed using iDiscover  
305 5.3 software. The data outputs derived from the automated mineral analysis

306 used in this study are: modal mineralogy, mean mineral particle size and  
307 QEMSCAN mineral and phase composition particle maps.

308

## 309 **Results**

310

311 In the following description the mineral abundance is referred to as  
312 major >10%, minor 1-10% and trace <1% abundance in the measured  
313 sample.

314

### 315 *Soil mineralogy variation*

316

317 The variation in the modal mineralogy for the 35 different sampling  
318 locations across SW England is shown in Figs. 3a – f, and tabulated in Table  
319 4. Although there is a range in bedrock geology in the study area the majority  
320 of the soil samples analysed are dominated by the same principal mineral  
321 types: quartz, plagioclase, K-feldspar, muscovite, biotite, chlorite, kaolinite, Fe  
322 silicates, Mg silicates, hornblende and tourmaline (Fig. 3a, c and e, Table 4).  
323 Less abundant minerals present are: epidote group minerals, sillimanite,  
324 topaz, other silicates, calcite, rutile, ilmenite, Fe-Mn oxides, chromite, apatite,  
325 zircon, xenotime, Ce phosphates, Fe/Cu sulphides, gypsum and “others” (Fig.  
326 3b, d, and f, Table 4). However, although the same major/minor and trace  
327 minerals tend to co-occur throughout the samples analysed, their relative  
328 abundance varies considerably (Fig. 3a). When the data are combined by the  
329 location rock type as described in Table 1, differences both between and  
330 within these groups are observed. Although there is some mineralogical  
331 variation within each lithological group, there is greater variation in modal  
332 mineralogy between the different lithological groupings than observed within  
333 an individual group. These differences between lithological groups indicate the  
334 potential for the ‘type’ of geology to be identified in an unknown sample. The  
335 observed difference detected between samples of the same lithology  
336 highlights that, within the study area, there is potential for individual locations  
337 to be identified by their particular signatures.

338 In addition to the modal mineralogical dataset, QEMSCAN particle  
339 compositional images provide further means of classifying differences

340 between the analysed soils. These reveal textural relationships between  
341 different mineral phases in the samples relating to grain size and mineral  
342 association, which can vary depending on the bedrock lithology on which the  
343 soil was developed. Representative particle images for selected soil samples  
344 developed on the different bedrock groups are shown in Fig. 4. Clear  
345 differences are observed in the mineral textures for soils developed on  
346 different rock types. For example, soils underlain by granitic bedrock include  
347 coarse grains of quartz, feldspar and mica minerals, whereas soil particles  
348 derived from metasedimentary rocks are highly heterogeneous, principally  
349 comprising grains of quartz intermixed with fine-grained clay minerals. No two  
350 soil samples analysed contain the same mineral types with the same relative  
351 abundance, grain size and texture, reflecting the distinctive nature of soil  
352 composition at different locations. The differences in characteristics between  
353 soils developed on different underlying bedrock, which are evident in  
354 groupings of automated mineralogy data (Fig. 3), could allow broad areas to  
355 be identified or ruled out by identifying the likely source lithologies from an  
356 unknown sample.

357

#### 358 *Soils grouped by bedrock lithology*

359

360 To assess the mineralogical variation throughout the study area in  
361 more detail the data for the same, or similar, bedrock geology is compared.  
362 Identification of differences in soil mineralogy between samples arising from  
363 similar bedrock geologies is required to define the range of compositions  
364 which could be indicative of a particular lithology. If specific differences are  
365 present, these could also be used to further refine the spatial scale of a  
366 geolocation assessment, such as identifying a particular group out of several  
367 candidate units within a region.

368

369 *Granites.* Six soil samples developed upon granite bedrock were analysed.  
370 These soils were identified as being derived from an underlying granitic  
371 geology by the consistent abundance of large (~100 µm) grains of quartz,  
372 plagioclase, K feldspar, biotite and muscovite (e.g. Fig. 4). The Cornubian  
373 granites have been subdivided into five mineralogical types and individual

374 named intrusions are commonly composite with more than one granite type  
375 present (Simons et al., 2016). Each of the soils sampled had distinctive  
376 compositions in minor and trace mineralogy, which were consistent with at  
377 least one of the characteristics of the underlying subcrop. This is indicative of  
378 the known spatial heterogeneity of the individual granite bodies (Simons et al.,  
379 2016).

380 Key mineralogical parameters and the known underlying granite types  
381 are presented in Table 5. The relative abundance of biotite and muscovite  
382 was indicative of biotite granite (G3, biotite > muscovite) underlying the Lands  
383 End granite soil, and two-mica granite (G1, biotite  $\approx$  muscovite) underlying the  
384 Tregonning-Godolphin and Carnmenellis Granite soils. High relative  
385 abundance of tourmaline in the St Austell and Dartmoor samples was  
386 indicative of G4 tourmaline granites in these sample areas. The Bodmin Moor  
387 Granite was characterised by an increased abundance of muscovite relative  
388 to biotite, consistent with muscovite granite (G2), and the abundance of  
389 'topaz' and 'sillimanite' (suggestive of the accessory mineral andalusite  
390 (Simons et al., 2016), which would be grouped with sillimanite during  
391 QEMSCAN analysis) was distinctive in this sample. Other mineralogical  
392 signatures in the group included kaolinite abundance, which was particularly  
393 high in the St Austell Granite soil. Replacement of feldspar by kaolinite is  
394 present in all of the granites of SW England, however, the St Austell Granite  
395 exhibits the most extensive kaolinisation (Ellis and Scott, 2004), and the high  
396 abundance in this sample is evidence that this mineralogical signature is  
397 preserved in the soil.

398 Within the study area, soils developed on granites possess both  
399 common overall characteristics that allows the underlying lithology to be  
400 identified, but also sufficient variety between samples to permit individual  
401 subcrops to be distinguished. Links between the composition of a soil and  
402 published characterisation of the granite type (Ellis and Scott, 2004; Simons et  
403 al., 2016) were possible, such that key mineral signatures can be used to rule  
404 out one or more of the subcrops as an unlikely source of the soil. This  
405 approach is powerful, but may be highly limited to locations or rock types  
406 which display this degree of mineralogical variation.

407

408 *Mafic igneous rock.* Six soil samples were analysed from locations where the  
409 reported bedrock geology was gabbro, dolerite or basalt (Table 3), and were  
410 grouped together as 'mafic igneous' rock types. Modal mineralogy data (Fig  
411 3a and 2b) shows that although there is some variation between these  
412 samples, they form a consistent grouping characterised by high abundance of  
413 chlorite and the presence of Mg silicates, both of which are indicative of  
414 altered mafic minerals.

415 Soils developed on dolerites and gabbros had similar overall  
416 composition and texture (e.g. Fig 4), comprising several species of coarse  
417 mono-mineralic grains suggestive of an igneous basement rock. The samples  
418 are however, subtly different to each other, with the gabbro derived soils  
419 containing more abundant K feldspar, hornblende and kaolinite, and less  
420 abundant Mg silicates, chlorite and Fe silicates than the dolerites. Highly  
421 specific mineral signatures were detected in some samples, in particular the  
422 presence of abundant hornblende in Sample 15, which within the study area  
423 was distinctive of the Trelan and Crousa Gabbro, part of the Lizard Complex.

424 The two soil samples from locations underlain by either basaltic  
425 calcareous tuffs of the Tintagel Volcanic Formation or the basalts-mudstones  
426 of the Milton Abbott Formation were distinct from the soils derived from  
427 dolerite/gabbro bedrock. In general, the soils associated with basaltic  
428 lithologies contain less abundant plagioclase, hornblende and tourmaline and  
429 more abundant K feldspar and biotite, although it should be noted that the  
430 basaltic lithologies are interbedded with mudstones. In addition, whilst the  
431 Tintagel Volcanic Formation is described as comprising calcareous basaltic  
432 tuffs, very few (0.10%) grains in the soil sample derived from this unit reported  
433 to the calcite mineral grouping. Geochemical and mineralogical studies have  
434 indicated that the Tintagel Volcanic Formation comprises both calcite-rich but  
435 also calcite-poor lithologies (Rice-Birchall and Floyd, 1988), hence it is  
436 possible that the sample collected was from soils developed on a calcite-poor  
437 lithological unit.

438 The soils underlain by mafic-igneous lithologies were found to form a  
439 consistent grouping of modal mineralogy and texture. Owing to the range of  
440 rock types grouped together, aspects of the mineralogy of both individual  
441 samples and rock types (e.g. gabbro vs dolerite) were found to be distinctive.

442 In contrast to granitic derived soils, the mineralogy of these samples is more  
443 strongly controlled by alteration products of mafic minerals, rather than the  
444 persistence of lithic fragments.

445

446 *Metamorphic rocks of the Lizard Complex.* Four soil samples collected from  
447 locations underlain by metamorphic rocks were analysed, all associated with  
448 the Lizard Complex (Fig. 1). It should be noted that this grouping of soil  
449 samples is rather arbitrary, as it does not necessarily represent a  
450 homogenous compositional grouping but could include rocks of differing  
451 mineralogy depending on protolith composition and metamorphic grade.  
452 Some of the samples showed similar major mineralogy to locations within the  
453 mafic-igneous group, however, they were identified as metamorphic derived  
454 soils by the enhanced abundance of mineral groups such as hornblende,  
455 epidote and Mg-silicates and a lower abundance of rutile and ilmenite.

456 Trends relating to specific units within the Lizard Complex were  
457 identified. Soil sample 14 has the highest abundance of hornblende (12.8%)  
458 and epidote (1.6%) of the entire sample set. Within the sample area this  
459 combination of hornblende and epidote, a common greenschist-facies  
460 metamorphic mineral, is distinctive of the Traboe Hornblende Schist  
461 underlying this sample location.

462 The abundance of Mg-silicate minerals was expected to be a  
463 distinguishing characteristic of soils underlain by the Lizard Serpentinite, as  
464 this is the compositional grouping which includes serpentine-group minerals  
465 (Table 3). However, although the soil from this area (Sample 16) contained  
466 1.4% Mg-silicates (serpentine group minerals), two samples from other areas  
467 contained higher abundances; samples 4 (11.1% Mg-silicates, gneiss) and 12  
468 (3.9% Mg-silicates, dolerite). The published geological map indicated that  
469 sampling location 4 was underlain by the Kennack Gneiss. However, this  
470 lithology has a patchy / localised distribution with pods of granitic gneiss  
471 surrounded by serpentinite. Consequently, the soil mineralogy data may be  
472 interpreted to suggest that either: (a) the published map is incorrect, and that  
473 the bedrock geology at this location is actually serpentinite, or that (b)  
474 periglacial processes have introduced serpentinite derived surficial deposits at  
475 this location. It should however, also be noted that both the Kennack Gneiss

476 (Sample 4) and Nare Head Dolerite (Sample 12) contain amphibole group  
477 minerals (Barnes et al., 1979), which (other than hornblende) would be  
478 characterised under the Mg silicate QEMSCAN grouping. Within the study  
479 area, soils with abundant Mg-silicate (>1%) were consistent with several  
480 underlying lithologies, however, this was highly distinctive, relating to three out  
481 of the thirty five locations sampled. More generally, this finding demonstrates  
482 the importance of obtaining detailed information on the composition of  
483 candidate lithologies to check for consistency with observed soil mineralogy.

484 Reference to regional soil geochemistry surveys revealed that a  
485 characteristic feature of the soils present on the Lizard Complex is a high  
486 relative abundance of chromium (Cr) (BGS, 2015). This was supported in the  
487 modal mineralogy data with elevated abundances of the mineral chrome  
488 spinel (Table 2) for soils developed on the Lizard Serpentinite (Sample 16)  
489 and also at sample location 4. The abundance of chrome spinel along with  
490 the serpentine minerals would support the interpretation that sampling location  
491 4 is not underlain by the Kennack Gneiss as indicated by the published map,  
492 but instead is underlain by serpentinite. This therefore provides a highly  
493 location specific signature which is absent in soils developed on chromium-  
494 poor rocks elsewhere in the sample region.

495

496 *Metamorphosed mudstone.* The dominant geological unit throughout the  
497 study area is Devonian low-grade metamorphosed mudstones and  
498 sandstones, the original deposition of which was strongly controlled by  
499 tectonics, within a series of separate depositional basins in the study area;  
500 Gramscatho, Looe, South Devon and Tavy (Fig. 1) (Leveridge, 2011;  
501 Leveridge and Shail, 2011b).

502 Thirteen soil samples developed upon Devonian metamorphosed  
503 mudstones were analysed, covering each of the depositional basins (Fig. 3c,  
504 d). These samples had a characteristic fine-grained texture (Fig. 4) and  
505 similar modal mineralogy, being dominated by major quartz, K feldspar,  
506 muscovite, biotite and chlorite along with minor/trace plagioclase, kaolinite  
507 and tourmaline. This combination of features permitted distinction of these  
508 samples as being derived from a metasedimentary source.

509 Despite the similar overall characteristics, it is clear that there are  
510 mineralogical variations within the dataset related to depositional basin (Fig.  
511 3c, d). For example, soil samples collected from locations up to ~60 km apart  
512 across the South Devon Basin (19, 23, 28 and 29) are all quartz-poor (less  
513 than 20% quartz, Fig. 3c), whilst those collected from the Gramscatho Basin  
514 (13, 18, 24, 26), again up to 60 km apart, have a consistently higher quartz  
515 abundance (>20-30%). The changing relative abundance of quartz may  
516 reflect differences in soil particle size, as quartz typically comprises larger  
517 sand/silt sized particles. However, the average particle size for Gramscatho  
518 Basin samples was finer than in the South Devon Basin samples, and  
519 therefore the quartz variations are likely to be representative of mineralogical  
520 differences between the locations rather than particle size.

521 In the soil mineralogy data, samples collected from the Trevoise Slate  
522 (Sample 19, Devon Basin) and the Tredorn Slate (Sample 21, Tavy Basin)  
523 formations both have elevated levels of chrome spinel (Table 2). Although  
524 this is not described in regional scale geological mapping, published soil  
525 geochemical surveys show an elevated chromium anomaly in these regions  
526 (BGS, 2015) consistent with the measured mineralogical data. Whilst Cr-rich  
527 soils are observed in other regions of the study area such as the Lizard  
528 Complex, a soil with meta-mudstone characteristics and the presence of Cr-  
529 spinel as a minor mineral indicates a restricted provenance to the Trevoise-  
530 Tredorn formations.

531

532 *Metamorphosed sandstone-mudstone.* Three soil samples were analysed  
533 from locations underlain by either metamorphosed Devonian sandstones or  
534 interbedded sandstones / mudstones (Figs. 3e and f). These were  
535 differentiated from the metamorphosed Devonian mudstones principally by  
536 texture; soils developed on lithologies with a sandstone component contained  
537 quartz as both sand-silt grade monominerallic and sand-grade polyminerallic  
538 grains (Fig. 5). The abundance of quartz is another key differentiator. Sample  
539 9, which was taken from a sandstone bedrock location has very high quartz  
540 abundance (73.7%), whilst soils from interbedded locations (samples 11 and  
541 25) are intermediate in quartz abundance between sandstone and mudstone  
542 locations. In these cases, the higher abundance of quartz is indicative of the

543 coarser grain size of the underlying rock at these locations, which along with  
544 grain texture, may allow the identification of soils developed on different grain  
545 size sedimentary or metasedimentary rocks.

546

547 *Carboniferous sedimentary rocks.* Sedimentary rocks of Carboniferous age  
548 which have undergone Variscan deformation and very low grade regional  
549 metamorphism occur within the eastern part of the study area. Three soil  
550 samples developed on Carboniferous mudstones, interbedded mudstones  
551 and sandstones, and chert were analysed (Fig. 3e and f). Soils developed on  
552 locations underlain by Carboniferous mudstone and chert formations  
553 (Samples 30 and 31) had similar fine-grained textures to the Devonian  
554 metamorphosed mudstone samples (e.g. Fig. 4 d and f). However, the lower  
555 metamorphic grade of the Carboniferous rock resulted in mineralogical  
556 differences, with lower biotite and muscovite (illite) abundances and more  
557 abundant chlorite, plagioclase and quartz reflecting lower thermal maturity of  
558 these bedrocks. The abundance of quartz in the 'chert' sample was not as  
559 high as may be expected, and therefore may arise from another interbedded  
560 component (e.g. mudstone) of the heterogeneous Teign (Newton) Chert  
561 Formation.

562 The sandstone-mudstone nature of Sample 33 could be readily  
563 identified amongst the Carboniferous sedimentary samples by the high  
564 abundance of quartz and coarse grain size. However, it could not be readily  
565 distinguished from the Devonian metamorphosed location (Sample 9); unlike  
566 the mudstones, little mineralogical change would be expected during the low  
567 grade metamorphism of a quartz-rich sandstone and therefore no difference  
568 between these samples was detected. This finding highlights that using  
569 automated mineralogy alone it may be difficult to distinguish between lower  
570 grade metamorphosed sediments and 'pure' sedimentary lithologies, and in  
571 areas predominantly composed of such rocks a wider suite of techniques will  
572 be required.

573

574 *Variation within a single location*

575

576 To examine the spatial variation at a more local scale in detail, one  
577 sample was selected at random and all five sampling points along a 0 to 250  
578 m transect were prepared for analysis. The modal mineralogy for these five  
579 samples, along with the replicate sample analysed from the first sampling  
580 point are illustrated in Fig. 6 and the data are presented in Table 6. The  
581 mineralogical data show the closest correspondence between the two  
582 replicate samples, but the overall suite of five samples collected over a  
583 distance of 250 m within the same agricultural field are a closely comparable  
584 data set. The data for the five samples along the individual transect show  
585 lower variance than the samples analysed from the different sampling  
586 locations. Thus if the data for the 35 separate sampling locations across the  
587 region are compared with the data for these samples, then it is clear that the  
588 soil modal mineralogical data would allow the identification of the correct  
589 sampling location. This supports the modal mineralogy data from the whole  
590 sample suite, which indicates that within the study area, the underlying  
591 bedrock geology is the dominant control on soil mineral composition.

592

## 593 **Discussion**

594

595 For forensic geolocation based on a soil sample, the ideal scenario  
596 would be to have access to representative samples from all the regions of  
597 interest to allow comparisons to be drawn directly. However, this is often not  
598 practical, and may be impossible in wide-area search investigations, which  
599 would be characteristic in nuclear forensics. Therefore, it is necessary to rely  
600 on comparison of measurable properties of an unknown soil with reference  
601 datasets to establish the potential provenance of a sample.

602 Most spatial soil surveys typically describe parameters such as grain  
603 size, organic content, pH, colour etc, which for geolocation purposes are not  
604 sufficiently descriptive. For example, soil surveys from the study area show  
605 little variation other than the major contrasts between upland peat soils versus  
606 the predominantly managed arable and grazing land. This contrasts markedly  
607 with the clear differences in soil mineralogy and particle textures revealed by  
608 automated mineralogical analysis, and this variety indicates that, within the

609 studied area, soil mineralogy is a more distinctive indicator of location than the  
610 overall soil classification or texture.

611 There are few comprehensive global datasets for soil mineralogy. The  
612 United States Geological Survey has an open access database for soil  
613 mineralogy of the USA, comprising 4857 samples, which equates to a  
614 sampling density of 1 sample per 1600 km<sup>2</sup> (Smith et al., 2014). As such this  
615 shows regional trends, but is of less value for forensic investigations. In the  
616 absence of comprehensive datasets for detailed soil mineralogy, other  
617 reference datasets must be used to predict locations at which a particular soil  
618 mineralogy may be found. In this paper we have demonstrated that, for the  
619 study area, soil mineralogy is consistent with the local underlying geology as  
620 represented by widely available regional scale geological mapping. Local  
621 scale geochemical surveys (BGS, 2015) can add an extra level of detail, such  
622 as the highly localised distribution of chromium around the Trevoise and  
623 Tredorn Slate formation locations, which was detected as a high relative  
624 abundance of chrome-spinel in these soils samples.

625 The nature of the correlation between bedrock composition and soil  
626 mineralogy will depend on a number of factors specific to the study area. In  
627 localities dominated by physical weathering, with typically arid to semi-arid  
628 climates, soils are more likely to be composed of liberated relic minerals grain  
629 and lithological fragments, and it is expected that there will be a strong  
630 correlation between the mineralogical composition of soils and the near-  
631 surface geology. However, in areas where chemical weathering  
632 predominates, and/or where soil profiles relate to past intervals of geological  
633 weathering, soil mineralogy may be substantially altered from the parent rock.  
634 Owing to the oceanic climate, soil formation in the study area is dominated by  
635 chemical weathering and hence is more representative of the second  
636 condition.

637 Soil mineralogy was also sufficient to distinguish individual occurrences  
638 of most of the lithology types within the study area. Consideration of the minor  
639 phases present in the granites showed differences related to the specific gran-  
640 ite type whereas the mineralogy of metasediment samples was shown to differ  
641 across different depositional basins. The extent to which this might be used to  
642 determine a specific location, rather than just distinguish two dissimilar sam-

643 ples, was varied and depended on the mineralogical variety within the parent  
644 rock. It was only in the case of the sandstone samples 9 and 33 that these  
645 distinctions could not be made; including between different metamorphosed  
646 grades of this lithology. Nevertheless, these results highlight the potential of a  
647 two-stage approach to predictive geolocation, whereby a generic lithology  
648 may be identified in the first instance, and, in favourable locations, further re-  
649 finement made by considering the details of local variations in specific occur-  
650 rences of that lithology. Whilst regional scale mapping is probably sufficient for  
651 the first aspect, more detailed data sources, such as specific publications,  
652 may be required for the second.

653 In the context of nuclear forensics, the capability provided by soil  
654 analysis to identify even the generic geological background may be of  
655 particular value. In many cases, such data may permit some potential  
656 locations to be excluded, rather than indicating an individual specific target  
657 location. The significance of this is that during a nuclear forensic  
658 investigation, the nature of a material being examined may indicate that it  
659 could only have been derived from a limited number of potential installations.  
660 Consideration of geolocation evidence (where present) may therefore allow  
661 some of these candidate sources to be ruled out based on their environmental  
662 setting, substantially narrowing the scope of the investigation. Further work to  
663 consider the collection and analysis of this type of evidence in nuclear forensic  
664 scenarios is needed. Although it may not be expected to recover soil traces in  
665 all cases, the power of this analysis to distinguish locations, if soil is present,  
666 highlights its value to these complex investigations.

667

## 668 **Conclusions**

669

670 In this study we aimed to test the extent to which soil mineralogy can  
671 be used to predict an underlying geological rock type, and how distinctive this  
672 may be of an individual unit of that lithology. The study area in SW England  
673 may represent a 'worst-case' type environment for this technique, as soils in  
674 the area are highly chemically altered from the host rock mineralogy through  
675 chemical weathering and human activity. The repetition of many of the major  
676 rock types throughout the area also challenged the capability of this analysis

677 to distinguish different locations with similar underlying rock types. In all  
678 sampled locations, signatures of the underlying bedrock geology were  
679 detected in the soil mineralogy and particle textures, and in 33 out of the 35  
680 samples, location-specific were detected permitting different units of the same  
681 major rock type to be distinguished.

682 This type of analysis is a powerful method for the identification of  
683 potential underlying bedrock units from soil samples, which can be used with  
684 readily available regional scale geological mapping to identify, and just as  
685 importantly rule out, candidate regions based on the known geology. The  
686 importance of this information will depend on the extent of geological variation  
687 within the area of investigation; if the entire area is underlain by the same  
688 lithology then other geolocation indicators may be more valuable.

689 In our area of interest in nuclear forensics, the provenance of a found  
690 nuclear material may be already somewhat constrained by known holdings  
691 and operations, and therefore identification of the geology type may allow  
692 some potential sources to be ruled out. Challenges relating to transfer and  
693 collection of soil evidence in these scenarios must be evaluated, however,  
694 consideration of environmental signatures such as soil evidence has the  
695 potential to provide a unique insight and perspective in these complex  
696 investigations.

697

## 698 **Acknowledgements**

699

700 We thank Holly Campbell (Helford Geoscience LLP) for assistance with the  
701 soil sampling and preparation of samples for analysis. Figure 1B reproduced  
702 from R.K. Shail and B.E. Leveridge. The Rhenohercynian passive margin of  
703 SW England: Development, inversion and extensional reactivation. *Comptes*  
704 *Rendus Geoscience*, 2009, 341 (issue2-3), 140-155. Copyright (c) 2009  
705 published by Elsevier Masson SAS. All rights reserved. Funding for this  
706 programme of research was provided by AWE plc to Helford Geoscience LLP.

707

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846

847

848 **Figure captions**

849

850 **Fig. 1.** Simplified maps showing distribution of sampling sites (A) and major  
851 near-surface geological features of SW England (B). Map B shows the main  
852 Devonian sedimentary basins (Gramscatho, Looe, South Devon and Tavy  
853 basins) along with the Lizard Ophiolite Complex (purple), the Start Complex  
854 and the exposed granites making up the Cornubian Batholith. Map A based  
855 on a Digimap extract © Crown Copyright and Database Right [June 2018].  
856 Ordnance Survey (Digimap Licence). Map B from Shail and Leveridge  
857 (2009).

858

859 **Fig. 2.** Example images for soil sampling location 1, near Halwyn Farm,  
860 Mousehole, West Cornwall, underlain by the Lands End Granite. Five soil  
861 samples were collected from the managed grassland field (A) at sampling  
862 positions of 0 (1/1), 10 (1/2), 50 (1/3), 100 (1/4) and 250 m (1/5) along the  
863 transect (B). Soil samples were collected by inserting a clean, single use  
864 plastic vial into the soil to a depth of 1-2 cm (C). Map A based on a Digimap  
865 extract © Crown Copyright and Database Right [June 2018]. Ordnance  
866 Survey (Digimap Licence).

867

868 **Fig. 3.** Modal mineralogy of the soil samples collected throughout Cornwall  
869 and Devon, SW England, grouped by lithology according to Table 2. Charts  
870 A, C and E show relative abundance of major (>10%) and minor (1-10%)  
871 minerals, Charts B, D and F show relative abundance of minor (1-10%) and  
872 trace (<1%) minerals.

873

874 **Fig. 4.** Diagram showing the QEMSCAN false colour particle images for  
875 representative samples from the six major bedrock groups in SW England.  
876 (A) Sample 10/1 St Austell Granite, (B) Sample 12/1 Nare Head Dolerite, (C)  
877 Sample 14/1 Traboe Hornblende Schist (Traboe Cumulate Complex), Lizard  
878 Ophiolite Complex, (D) Sample 23/1 Devonian Polzeath Slate Formation  
879 metamorphosed mudstone, (E) Sample 9/1 Devonian Staddon Formation  
880 metamorphosed sandstone and (F) Sample 30/1 Carboniferous Teign  
881 (Newton) Chert Formation.

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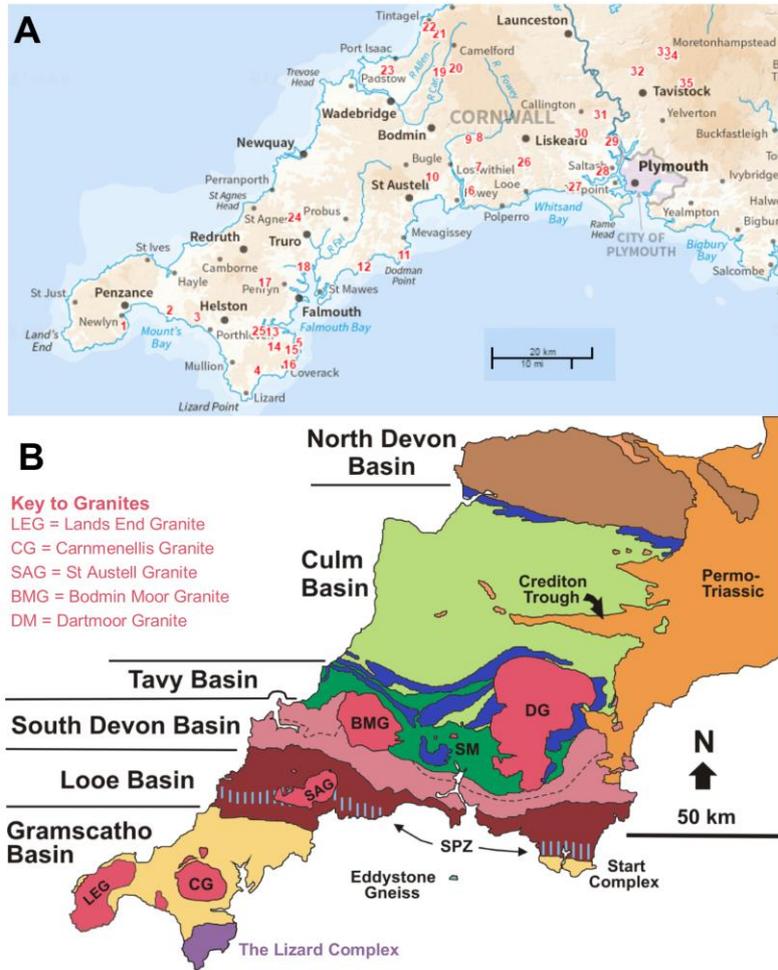
883 **Fig. 5.** Diagram showing the QEMSCAN false colour particle images for two  
884 soil samples developed on Devonian metasedimentary bedrock units from SW  
885 England. (A) Sample 9/1 Staddon Formation metamorphosed sandstones,  
886 (B) Sample 29/1 Tavy Formation metamorphosed mudstone. The quartz grain  
887 size and texture is a key discriminator of different sedimentary grades.

888

889 **Fig. 6.** Modal mineralogy of the 6 soil samples collected from sampling  
890 location 24 Porthtowan Formation bedrock, near Allet, Truro, Cornwall, along  
891 a 250 m transect. (A) Relative abundance of major (>10%) and minor (1-  
892 10%) minerals. (B) Relative abundance of minor (1-10%) and trace (<1%)  
893 minerals.

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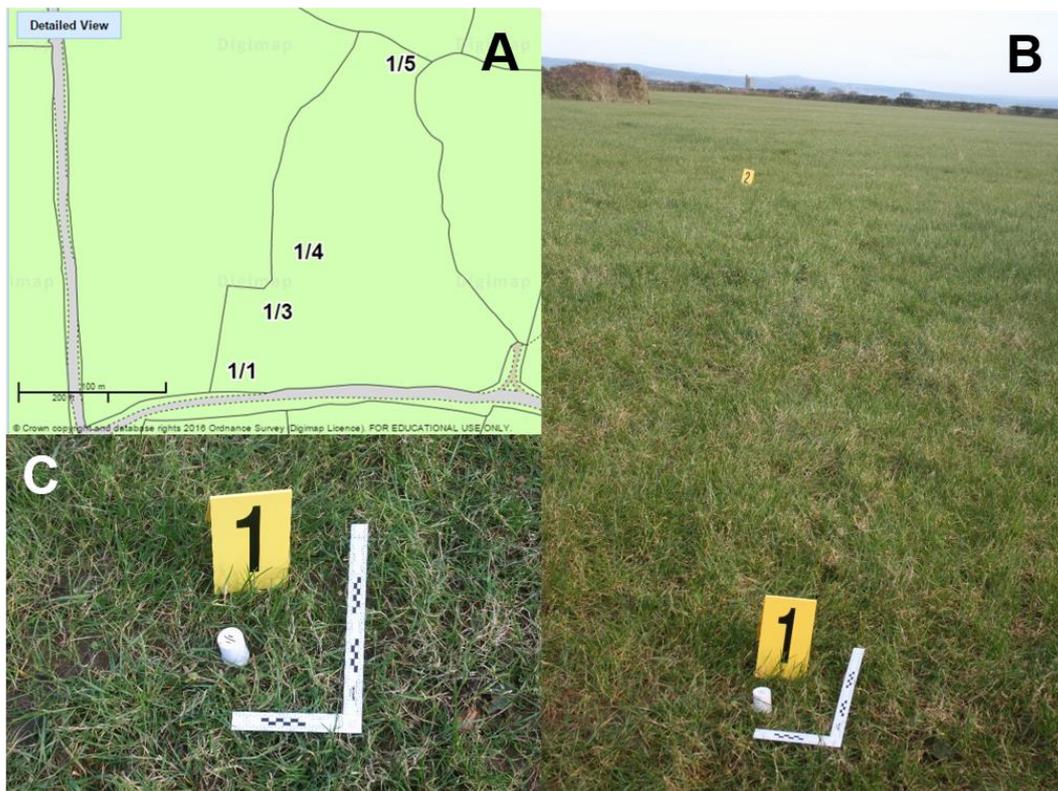
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897 Figure 1.

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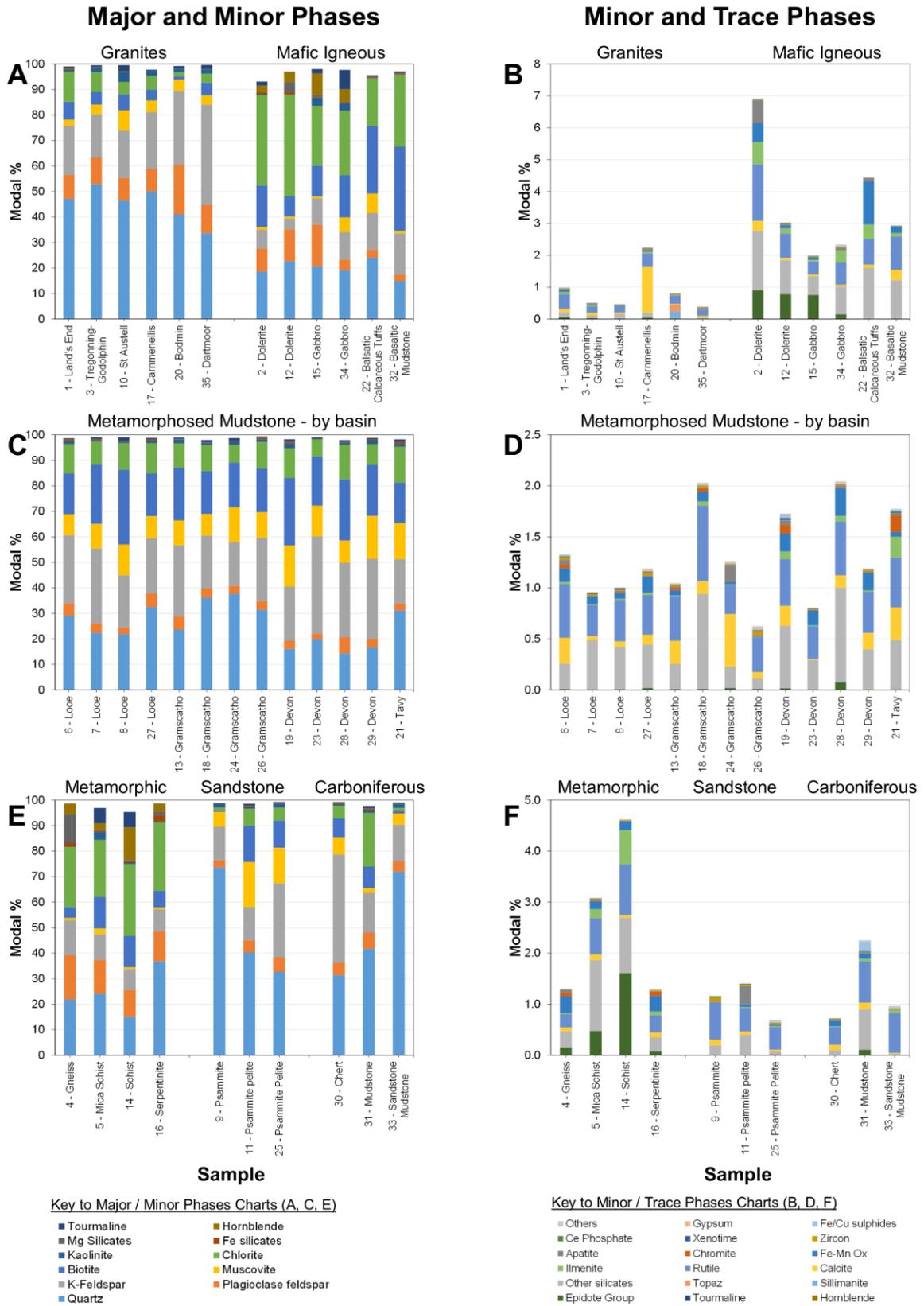


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901 Figure. 2

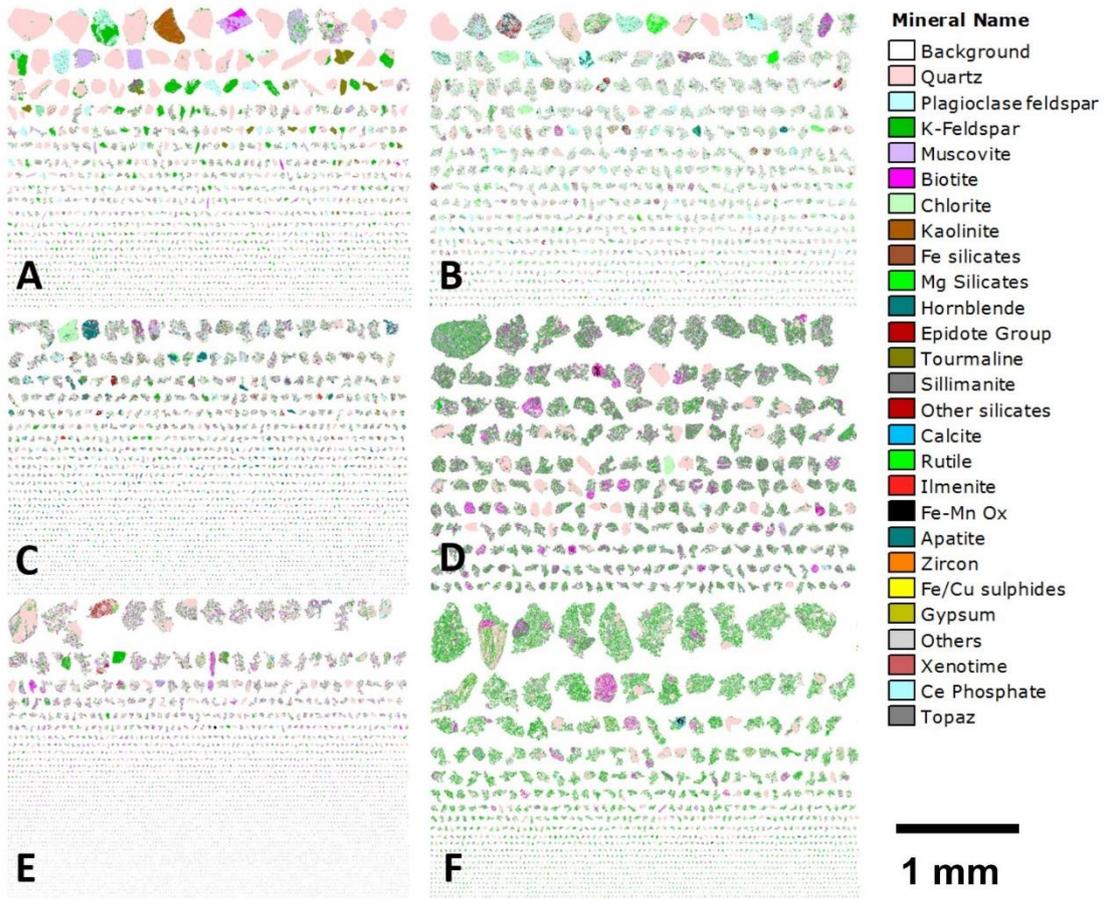
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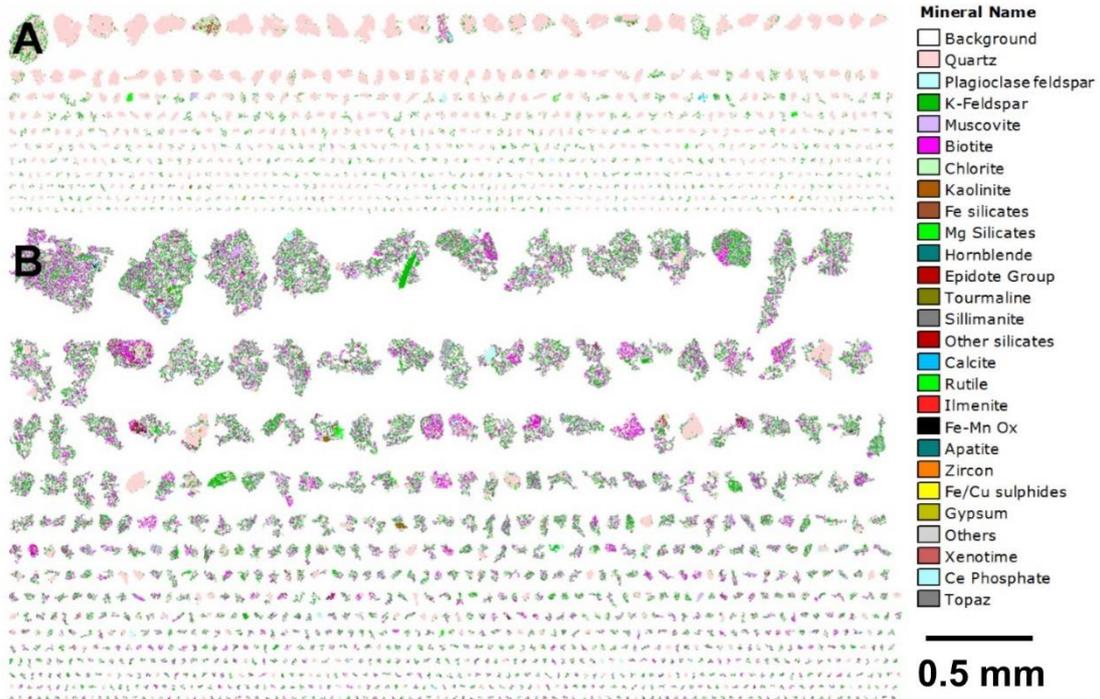
904 Figure 3

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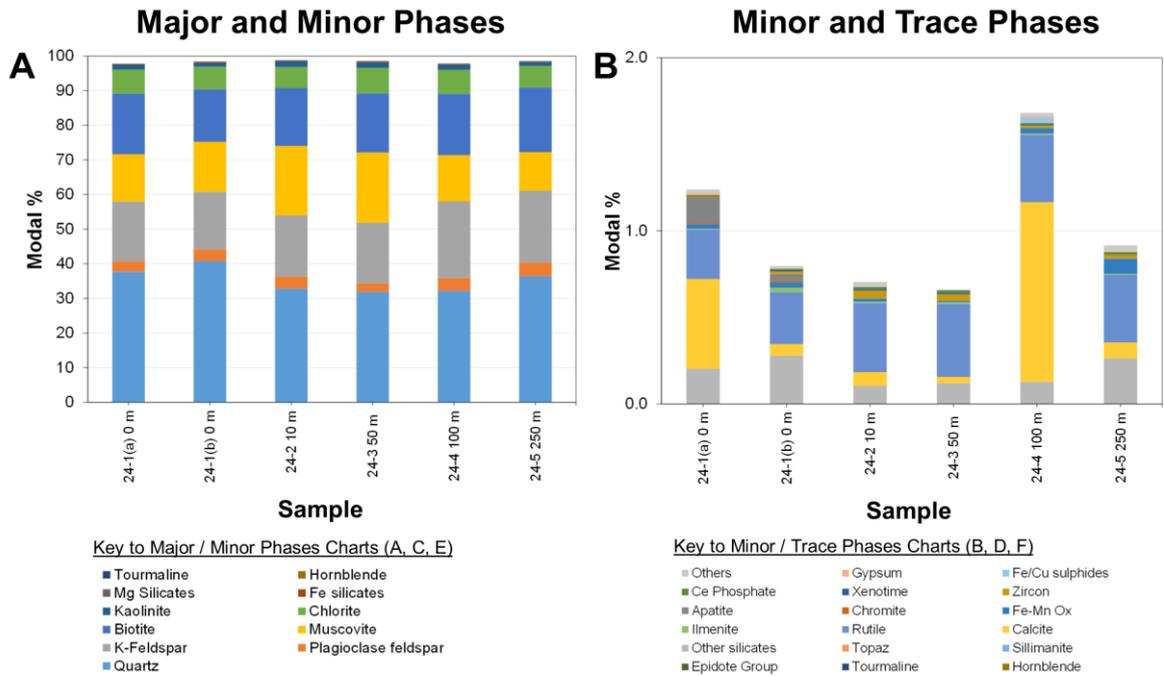
Figure 4.



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Figure 5

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Figure 6

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919 **Table 1.** *Sampling locations, bedrock geology, OS grid reference and land use.*

	Rock type	Formation	Age	Land use	Grid Reference
1/1	Granite	Lands End Granite	Permian	Resown grassland	SW45821 25934
2/1	Dolerite	Dolerite	Devonian	Pasture	SW54743 29115
3/1	Granite	Tregonning-Godolphin Granite	Permian	Managed grassland	SW60211 27618
4/1	Gneiss	Kennack Gneiss	Devonian (?Famennian)	Managed grassland	SW72090 16757
5/1	Mica schist	Old Lizard Head Formation	?Cambrian	Managed grassland	SW80076 22399
6/1	Meta-mudstone	Dartmouth Group	Devonian (Lochkovian-Pragian)	Pasture	SX14051 53330
7/1	Meta-mudstone	Meadfoot Group	Devonian (Pragian-Emsian)	Crops - maize	SX15377 58140
8/1	Meta-mudstone	Saltash Formation	Devonian-Carboniferous (Emsian-Tournasian)	Resown grassland	SX15571 63977
9/1	Meta-sandstone	Staddon Formation	Devonian (Emsian)	Pasture (very wet)	SX13368 63620
10/1	Granite	St Austell Granite	Permian-Carboniferous	Crops - maize	SX05528 56036
11/1	Meta-sandstone / mudstone	Dodman Formation	Early Devonian	Pasture	SX00095 40220
12/1	Dolerite	Nare Head Dolerite	Devonian (Givetian-Frasnian)	Managed grassland	SW92165 37879
13/1	Meta-mudstone	Roseland Breccia Formation	Devonian (Givetian-Frasnian)	Managed grassland (wet)	SW74187 24719
14/1	Schist	Traboe Hornblende Schist (Traboe Cumulate Complex)	Devonian (?Emsian)	Managed grassland	SW 74614 21750
15/1	Gabbro	Trelan and Crousa Gabbro	Devonian	Managed grassland	SW77884 21082
16/1	Serpentinite	Lizard Serpentinite	Devonian (Emsian) (mantle exhumation)	Managed grassland	SW77486 18004
17/1	Granite	Carmmenellis Granite	Permian	Managed grassland	SW72821 34663
18/1	Meta-mudstone	Mylor Slate Formation	Devonian (Frasnian-Famennian)	Managed grassland	SW80249 37757
19/1	Meta-mudstone	Trevoze Slate Formation	Devonian (Frasnian-Famennian)	Managed grassland	SX07049 77239
20/1	Granite	Bodmin Granite	Permian	Moorland (grazed)	SX10137 78066
21/1	Meta-mudstone	Tredorn Slate Formation	Devonian (Famennian)	Managed grassland	SX07053 85000
22/1	Basaltic calcareous tuffs	Tintagel Volcanic Formation	Carboniferous (Visean)	Coastal grassland	SX04879 86451
23/1	Meta-mudstone	Polzeath Slate Formation	Devonian (Frasnian-Famennian)	Managed grassland	SW96717 77740
24/1	Meta-mudstone	Porthtowan Formation	Devonian (Eifelian-Frasnian)	Grassland margin to cultivated field	SW78531 47936
25/1	Meta-sandstone / mudstone	Portscatho Formation	Devonian (Givetian-Frasnian)	Managed grassland	SW71594 24780
26/1	Meta-mudstone	Bovisand Formation	Devonian (Pragian-Emsian)	Managed grassland	SX23501 58986
27/1	Meta-mudstone	Whitsand Bay Formation	Devonian (Lochkovian-Pragian)	Managed grassland	SX33649 54060
28/1	Meta-mudstone	Torpoint Formation	Devonian (Frasnian-Famennian)	Managed grassland	SX39086 57333
29/1	Meta-mudstone	Tavy Formation	Devonian (Frasnian-Famennian)	Grazed pastureland	SX40710 63197
30/1	Chert	Teign (Newton) Chert Formation	Carboniferous (Visean)	Grassland margin to cultivated field	SX34764 64914
31/1	Mudstone	Brendon Formation	Carboniferous (Visean)	Managed grassland	SX38587 68631
32/1	Basalt-Mudstone	Milton Abbott Formation	Carboniferous (Visean)	Managed grassland	SX45785 77497
33/1	Sandstone-Mudstone	Bealsmill Formation	Carboniferous (Serpukhovian-Bashkirian)	Moorland (near to Wheal Betsy mine)	SX50899 81429
34/1	Dolerite	Dolertie	Devonian-Carboniferous	Moorland (grazed)	SX52403 80699
35/1	Granite	Dartmoor Granite	Permian	Moorland	SX55354 75059

920 **Table 2.** *Soil sample numbers, laboratory codes, number of particles*  
 921 *measured and number of individual EDS analyses.*  
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Sample	Block Code	Number of particles	Number of EDS analyses
1/1	16HG31	5017	170581
2/1	16HG32	5087	340966
3/1	16HG33	5257	95018
4/1	16HG34	5159	183139
5/1	16HG35	5009	179889
6/1	16HG36	5348	144111
7/1	16HG37	5051	487685
8/1	16HG38	5065	344579
9/1	16HG39	5362	60501
10/1	16HG3A	5249	199123
11/1	16HG3B	5152	200560
12/1	16HG3C	5077	214346
13/1	16HG3D	5060	153689
14/1	16HG3E	5570	144240
15/1	16HG3F	5109	121620
16/1	16HG3G	5170	98969
17/1	16HG3H	5148	72579
18/1	16HG3I	5145	109501
19/1	16HG3J	5210	89146
20/1	16HG3K	5392	73202
21/1	16HG3L	5280	60674
22/1	16HG3M	5257	73762
23/1	16HG3N	5080	325097
24/1	16HG3O	5385	113463
25/1	16HG3P	5304	117203
26/1	16HG3Q	5021	102166
27/1	16HG3R	5105	112770
28/1	16HG3S	5191	172621
29/1	16HG3T	5197	135687
30/1	16HG3U	5087	197134
31/1	16HG3V	5645	62752
32/1	16HG3W	5524	110229
33/1	16HG3X	5075	50734
34/1	16HG3Y	5098	81299
35/1	16HG3Z	5220	77215
24/1	16HG3AA	5213	95367
24/2	16HG3AB	5057	67539
24/3	16HG3AC	5066	87886
24/4	16HG3AD	5075	119598
24/5	16HG3AE	5123	107509

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924 **Table 3.** *Mineral groupings used to process automated mineralogy data.*

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<b>Mineral Grouping</b>	<b>Description</b>
Quartz	Quartz. May include other silica minerals
Plagioclase feldspar	Plagioclase (albite to anorthite solid solution)
K feldspar	K feldspar such as orthoclase and microcline
Muscovite	Muscovite. May include alteration products after feldspars such as sericite. Illite also reports to this category.
Biotite	Biotite and phlogopite. Would also include glauconite
Chlorite	Chlorite. Man-made slags would also report to this category
Kaolinite	Kaolinite, halloysite, dickite
Fe silicates	Nontronite and other Fe silicates
Mg silicates	Mg silicates such as serpentine group minerals, orthopyroxene, olivine
Hornblende	Hornblende and other amphiboles
Tourmaline	Tourmaline group minerals
Epidote Group	Epidote group minerals
Sillimanite	Sillimanite, andalusite, kyanite
Topaz	Topaz
Other silicates	Garnet. Al silicates
Calcite	Calcite, ankerite, dolomite, magnesite
Rutile	Rutile, titanite (sphene)
Ilmenite	Ilmenite
Fe-Mn Ox	Fe oxides (magnetite, hematite), Mn oxides
Chromite	Chrome spinel
Apatite	Apatite, biogenic apatite
Zircon	Zircon
Xenotime	Xenotime
Ce phosphate	Monazite
Fe / Cu sulphides	Pyrite, chalcopyrite
Gypsum	Gypsum
Others	Any other mineral not reported above

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937 **Table 4.** *Modal mineralogical data for the 35 soil samples analysed throughout*  
 938 *Cornwall and Devon, SW England.*  
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	1-1	2-1	3-1	4-1	5-1	6-1
Quartz	47.17	18.57	52.89	21.80	24.17	29.10
Plagioclase feldspar	9.38	9.06	10.39	17.45	13.24	4.80
K-Feldspar	19.18	7.36	16.95	13.71	10.05	26.77
Muscovite	2.44	1.11	3.82	0.90	2.26	8.21
Biotite	7.01	16.26	5.03	4.26	12.40	15.93
Chlorite	11.83	35.42	7.74	23.50	22.30	11.51
Kaolinite	0.84	0.29	1.71	0.08	3.18	1.30
Fe silicates	0.44	0.80	0.27	1.63	0.61	0.67
Mg Silicates	0.02	0.12	0.00	11.05	0.11	0.01
Hornblende	0.19	2.56	0.01	4.31	2.62	0.02
Tourmaline	0.06	0.90	0.03	0.15	0.48	0.01
Epidote Group	0.51	1.52	0.65	0.02	5.97	0.34
Sillimanite	0.00	0.00	0.01	0.00	0.00	0.00
Topaz	0.01	0.00	0.01	0.00	0.00	0.00
Other silicates	0.15	1.86	0.07	0.31	1.39	0.25
Calcite	0.10	0.32	0.08	0.08	0.11	0.25
Rutile	0.45	1.77	0.19	0.26	0.70	0.52
Ilmenite	0.08	0.70	0.04	0.02	0.18	0.02
Fe-Mn Ox	0.08	0.60	0.01	0.32	0.15	0.13
Chromite	0.01	0.02	0.01	0.07	0.01	0.04
Apatite	0.00	0.69	0.01	0.06	0.01	0.04
Zircon	0.02	0.02	0.01	0.01	0.01	0.03
Xenotime	0.01	0.01	0.02	0.00	0.01	0.00
Ce Phosphate	0.00	0.00	0.00	0.00	0.00	0.00
Fe/Cu sulphides	0.01	0.00	0.00	0.00	0.00	0.01
Gypsum	0.00	0.01	0.00	0.00	0.00	0.00
Others	0.01	0.02	0.01	0.00	0.02	0.00

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955 **Table 5.** Key mineralogical parameters for the six granite derived soils compared  
 956 with the classification of the exposed granite types from (Simons et al., 2016).  
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	1 - Lands End Granite	3 - Tregonning Godolphin Granite	10 - St Austell Granite	17 - Cammenellis Granite	20 - Bodmin Moor Granite	35 - Dartmoor Granite
Kfsp	19.18	16.95	18.62	22.39	29.09	39.31
Plag	9.38	10.39	8.89	8.79	19.33	11.11
Biot	7.01	5.03	6.22	4.21	1.30	4.88
Musc	2.44	3.82	7.87	4.47	4.47	3.70
Kaol	0.84	1.71	3.93	1.79	1.30	1.67
Tourm	0.51	0.65	2.40	0.52	1.00	1.58
Sillim	0	0.01	0.01	0.03	0.23	0
Topaz	0.01	0.01	0	0.02	0.21	0
<b>Granite types</b>	G3 biotite granite G4 tourmaline granite	G1 two mica granite G5 topaz granite	G3 biotite granite G4 tourmaline granite	G1 two mica granite G2 Muscovite granite	G1 two mica granite G2 Muscovite granite	G3 biotite granite G4 tourmaline granite

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988 **Table 6.** *Modal mineralogical data for the soil samples collected along a 250 m*  
 989 *transect at Location 24, near Allet, Truro, Cornwall, with replicate sample 24/1.*  
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	24-1(a) 0 m	24-1(b) 0 m	24-2 10 m	24-3 50 m	24-4 100 m	24-5 250 m
Quartz	37.69	40.77	32.84	31.91	32.20	36.42
Plagioclase feldspar	3.06	3.41	3.48	2.41	3.68	3.91
K-Feldspar	17.22	16.60	17.68	17.51	22.13	20.75
Muscovite	13.65	14.45	20.03	20.31	13.37	11.17
Biotite	17.49	15.12	16.71	17.05	17.67	18.66
Chlorite	7.00	6.58	6.11	7.38	6.98	6.23
Kaolinite	1.40	1.15	1.70	1.66	1.54	1.17
Fe silicates	0.25	0.26	0.18	0.31	0.22	0.21
Mg Silicates	0.01	0.00	0.05	0.00	0.01	0.03
Hornblende	0.01	0.00	0.01	0.00	0.03	0.05
Tourmaline	0.97	0.85	0.46	0.79	0.45	0.47
Epidote Group	0.02	0.01	0.00	0.00	0.03	0.02
Sillimanite	0.00	0.00	0.01	0.00	0.00	0.00
Topaz	0.00	0.00	0.02	0.00	0.00	0.00
Other silicates	0.20	0.28	0.10	0.12	0.13	0.26
Calcite	0.52	0.07	0.08	0.04	1.04	0.09
Rutile	0.29	0.30	0.40	0.42	0.39	0.39
Ilmenite	0.01	0.03	0.01	0.01	0.01	0.01
Fe-Mn Ox	0.03	0.03	0.02	0.01	0.03	0.09
Chromite	0.01	0.01	0.00	0.00	0.00	0.00
Apatite	0.15	0.04	0.00	0.00	0.00	0.00
Zircon	0.01	0.01	0.04	0.04	0.02	0.02
Xenotime	0.00	0.01	0.01	0.01	0.01	0.00
Ce Phosphate	0.01	0.01	0.01	0.01	0.01	0.01
Fe/Cu sulphides	0.00	0.00	0.00	0.00	0.03	0.00
Gypsum	0.01	0.00	0.00	0.00	0.01	0.01
Others	0.02	0.01	0.03	0.00	0.02	0.03

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