Contrasting mechanisms for crustal sulphur contamination of mafic magma: evidence from dyke and sill complexes from the British Palaeogene Igneous Province

Hannah S. R. Hughes[1*], Adrian J. Boyce[2], Iain McDonald[1], Brett Davidheiser-Kroll[2], David A. Holwell[3], Alison McDonald[2], Anthony Oldroyd[1],

[1]School of Earth and Ocean Sciences, Cardiff University, Park Place, Cardiff, CF10 3AT, UK
[2]Scottish Universities Environment Research Centre, Rankine Avenue, East Kilbride, Glasgow, G75 0QF, UK
[3]Department of Geology, University of Leicester, University Road, Leicester, LE1 7RH, UK

*Corresponding author email: HughesH6@cf.ac.uk; Telephone: +44(0)29 208 76420

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3   **Abstract**

4   The addition of crustal sulphur to magma can trigger sulphide saturation: a process fundamental to

5   the development of some Ni-Cu-PGE deposits. In the British Palaeogene Igneous Province, mafic and

6   ultramafic magmas intrude a thick sedimentary sequence offering opportunities to elucidate

7   mechanisms of magma–crust interaction in a setting with heterogeneous S isotope signatures. We

8   present S-isotopic data from sills and dykes on the Isle of Skye. Sharp contrasts exist between

9   variably light δ^{34}S in Jurassic sedimentary sulphide (-35 to -10‰) and a local pristine magmatic δ^{34}S

10   signature of -2.3 ± 1.5‰. Flat-lying sills have restricted δ^{34}S (-5 to 0‰) while steeply dipping dykes

11   are more variable (-30 to -2‰). We suggest that the mechanism by which magma is intruded exerts

12   a fundamental control on the degree of crustal contamination by volatile elements. Turbulent flow

13   within narrow, steep magma conduits, discordant to sediments, and developed by brittle

14   extension/dilation have maximum contamination potential. In contrast, sill-like conduits emplaced

15   concordantly to sediments show little contamination by crustal S. The province is prospective for Ni-

16   Cu-PGE mineralization analogous to the sill-hosted Noril'sk deposit and Cu/Pd ratios of sills and

17   dykes on Skye indicate that magmas had already reached S-saturation before reaching the present

18   exposure level.

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20   Supplementary material: Supplementary Material (A) provides details of the whole-rock chemical

21   sulphur extraction methodology. Supplementary Material (B) provides simplified geological maps of

22   the Isle of Skye (Geographical Information System shapefiles © Edina Digimaps 05/10/2011), a

23   geological cross-section of part of southern Skye, and a stratigraphic log of the Mesoproterozoic and

24   Mesozoic sediments of western Scotland. Supplementary Material (C) provides trace element

25   diagrams of sills and dykes used in this study. Supplementary Material (D) provides tables for QA/QC

26   of S-isotope results. In addition, Table A gives sample location information, Table B gives whole-rock

27   major and trace element results for sill and dyke samples, and Table C provides whole-rock major

28   and trace element compositions for Jurassic mudrocks. All Supplementary Material is available at

29   www.geolsoc.org.uk/SUP00000

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32   Keywords: Sulphur isotopes, British Palaeogene Igneous Province, magma, crustal contamination

33   The Palaeogene North Atlantic Igneous Province (NAIP) formed from the intrusion and eruption of

34   mantle-sourced magmas, after the impingement of the proto-Icelandic mantle plume onto overlying

35   lithosphere. Continental rifting initiated in the Palaeogene, with an initial phase of magmatism at 62

36   Ma in the UK, Greenland and Baffin Island, and ultimately led to the opening of the Atlantic Ocean

37   (continental rifting initiated c. 55 Ma; Saunders *et al.*, 1997). The main products of this prolonged

38   period of magmatism were tholeiitic basalts, in addition to alkali basalts. Where the mantle plume

39   impinged on continental areas, magma fractionation, differentiation, and contamination were

40   commonplace.

41   The Isle of Skye and surrounding Western Isles of Scotland host a number of prominent intrusive and

42   volcanic features of the British Palaeogene Igneous Province (BPIP; Fig. 1a). On Skye, upper crustal

43   mafic and ultramafic intrusions were injected through Archaean Lewisian gneiss basement of the

44   North Atlantic Craton and Mesoproterozoic Torridonian sandstones, siltstones and mudstones. At

45   the current erosion level, the BPIP intrusions (for example the flat-lying Trotternish Sill Complex and

46   an extensive vertical suite of basalt dykes; Fig. 1a) have also penetrated through a thick succession of

47   Mesozoic sedimentary rocks of the Hebrides Basin, which are well exposed and can be

48   demonstrated to be physically contaminating BPIP magmas (e.g., via the presence of xenoliths).

49   The sulphur content of magma may be dramatically increased during crustal contamination and may

50   exceed the saturation concentration, triggering exsolution of an immiscible sulphide liquid into

51   which chalcophile elements such as Cu, Ni and the PGE may partition (e.g., Naldrett 2004, 2011 and

52   references therein). Magmas that intrude through the Jurassic mudrocks have particular potential

53   for Ni-Cu-PGE mineralization as these sediments would be expected to contain a high level of pyrite

54   with biogenically-derived sulphur (e.g., Fisher & Hudson, 1987). The source of this crustal sulphur

55   can be established by comparing the $\delta^{34}S$ signature of the magmas with that of the country rocks

56   through which it has ascended. In crustal sediments, a substantial range of $\delta^{34}S$ compositions may be

57   preserved in diagenetic sulphides, mostly pyrite. The sulphide isotopic range arises from mass-

58 dependent fractionation during bacterial reduction of seawater sulphate in the contemporaneous

59 diagenetic marine environment, which produces sulphide typically ≤20‰ lighter than the starting

60 sulphate (Ohmoto & Goldhaber, 1997). The range of δ^{34}S can range over 10's of per mille (‰), with

61 Palaeozoic and Mesozoic sulphides extending to more negative values than Precambrian rocks

62 (Canfield & Teske, 1996; Parnell *et al.,* 2010). By contrast, the δ^{34}S signature of the mantle usually

63 ranges from -2 to +2‰, and magmatic signatures can range from -4 to +4‰ depending on the

64 oxidation state of the magma (Ohmoto & Rye, 1979). Due to the dynamic nature of magma

65 intrusion, the crustal S-isotopic signature may be transported away from the immediate site of

66 contamination throughout magma ascent. Therefore crustal S input could be detected remotely

67 from concentrated sulphide mineralization. Such contamination is typically revealed by analysis of S-

68 isotopes in sulphide minerals or whole-rock samples. Our study uses and develops both techniques

69 to test for crustal contamination in the BPIP. The addition of crustal sulphur to a magmatic system is

70 commonly considered essential in the formation of orthomagmatic Ni-Cu-PGE mineralization (e.g.,

71 Naldrett 2004, 2011; Ripley & Li, 2013) and hence identifying the controls on crustal sulphur

72 contamination, and where this may be occurring will refine strategies for targeting of Ni-Cu-PGE

73 orthomagmatic mineralization in the BPIP and elsewhere.

74 We present the first published sulphur isotope compositions for the BPIP, determining a 'framework'

75 for the country rock δ^{34}S of the Hebrides Basin vs. δ^{34}S of the BPIP upper crustal intrusions on Skye

76 (including samples from the Lewisian basement, Torridonian sediments, and various Jurassic

77 sediments of the Hebrides Basin). We identify the 'background' plume magmatic sulphur isotopic

78 composition of the BPIP, and investigate if widespread crustal sulphur contamination of magmas

79 took place in NW Scotland. Coupled with whole-rock Ni, Cu and platinum group element (PGE)

80 concentrations, this contributes towards the development of a new model for magmatic sulphur

81 contamination, dependent on the orientation and energy by which magma is intruded through a

82 conduit, which impacts upon near-field exploration models for orthomagmatic Ni-Cu-PGE deposits.

83   *<< insert Fig. 1 >>*

84

1. Geology of the Hebridean portion of the British Palaeogene Igneous Province (BPIP)

86   The British Palaeogene Igneous Province (BPIP) is part of the earliest magmatic series of the NAIP,

87   which includes Palaeogene rocks of the Hebridean Igneous Province (along the W coast of Scotland)

88   and Northern Ireland (Fig. 1a). In Scotland, the BPIP includes the Isles of Mull, Skye, Arran, the Small

89   Isles (Rum, Eigg, Muck, Canna and Sanday), together with the mainland igneous complex of

90   Ardnamurchan and lava flows of Morvern. Thus, the BPIP in Scotland extends over a number of

91   tectonic terranes.

92   Crustal impingement of mantle-sourced mafic magmas resulted in subaerial eruptions along fissure-

93   type feeders (Kent *et al.*, 1998), now seen as a laterally continuous linear array of dyke swarms, with

94   individual dykes typically < 10m wide (Emeleus & Bell, 2005). These NW-SE trending dykes are

95   perpendicular to the North Atlantic rift margin and indicative of the contemporaneous NE-SW

96   directed extension of NW Europe at this time (Emeleus & Bell, 2005). Sills associated with lava fields

97   and central complexes normally occur within Mesozoic sediments below lavas, suggesting a

98   relatively shallow emplacement depth (probably < 1km). Individual sill thicknesses range from up to

99   10s of metres, amalgamating to form complexes 100s metres in thickness. Sills are often observed to

100   have their own compositional characteristics, unrelated to other overlying lavas. Sills predominantly

101   consist of alkali olivine basalts and tholeiitic basalts, although trachytes and rhyolites are recognised

102   as minor fractionation products (Emeleus & Bell, 2005).

1.1 Palaeogene geology of the Isle of Skye

104   The Isle of Skye records numerous exposures of BPIP magmatic rocks injecting through and into a

105   thick Mesozoic sedimentary sequence, part of the Hebrides Basin (Harker, 1904). Below the

106   Mesozoic rocks, a thick crustal pile of deformed Neoproterozoic (Torridon and Sleat Group)

107   sediments are present above Archaean (Lewisian) basement (Fig. 1b). Hence the Isle of Skye

108   provides an excellent opportunity to study the sources, extent and controls on crustal sulphur

109   contamination in a continental rift environment.

110   **1.1.1 The Trotternish Sill Complex, northern Skye**

111   The Trotternish Sill Complex outcrops on the Trotternish Peninsula on the north of the Isle of Skye

112   intruding Jurassic sandstones, limestones and marls (Fig. 1a; Gibb & Gibson, 1989). The picrites,

113   picrodolerites, and crinanites (analcite olivine dolerites) present throughout the sill complex are

114   genetically related, and thought to result from differentiation of an alkali-olivine basalt magma

115   (Gibson & Jones, 1991) with varying crustal contamination (Simkin, 1967; Gibson, 1990). This

116   horizontal sill complex post-dates the Skye Lava Group and is at least 250m thick, with individual sills

117   measuring 10's of metres in thickness. Rafts or 'packages' of Jurassic sediments (now baked) occur

118   sporadically within the thickness of the Trotternish Sill Complex in various locations. Sills can display

119   multiple and composite lithologies or melt generations, with banding commonplace (Gibson &

120   Jones, 1991; Emeleus & Bell, 2005). In these cases, the lower zones of a sill are the most mafic. For a

121   detailed 'stratigraphy' of the Trotternish Sill Complex, the reader is referred to Gibson (1990) and

122   Gibson & Jones (1991).

123

124   **1.1.2 Dykes from the Robustan area of the Strathaird Peninsula, southern Skye**

125   Dyke swarms on the Isle of Skye are predominantly directed in a NW-SE azimuth and caused

126   significant local crustal dilation. Most dykes are < 1 m thick, but some wider dykes (e.g., Strathaird

127   Peninsula) display evidence of incremental build-up due to multiple mafic magma injections,

128   resulting in magma mingling and layering within dykes (Platten, 2000). The Skye Dyke Swarm

129   intrudes both lavas and gabbros of the Cuillin Centre, and are mutually cut by cone-sheets (Bell &

130   Williamson, 2002). Compositionally, the Skye Dyke Swarm ranges from silicic pitchstones, to alkali

131   and tholeiitic basalts, and trachytes (Emeleus & Bell, 2005). Ultramafic dykes are predominantly

132   found closest to the Cuillin Central Complex.

133   At Robustan on the Strathaird Peninsula, southern Isle of Skye (see Fig. 1a), abundant NNW-SSE to

134   NW-SE trending basaltic dykes (hereafter called "Robustan dykes") cross-cut both the lavas and

135   sediments (see cross-section, Fig. 2). They represent part of a larger dyke swarm, formed and

136   intruded after the eruption of the Strathaird lavas, and after the intrusion of the Cuillin Central

137   Complexes to the N and NW. The cross-section in Figure 2 displays the transect from which dyke and

138   sediment samples were collected. Rare or minor examples of more evolved dyke compositions are

139   also observed, including basaltic trachyandesites. The dykes are typically vertical and range from 30

140   cm to 10 m in width. The dykes are finely crystalline (crystal size < 1mm) and equigranular. Chilled

141   margins are usually absent.

142   *<< insert Fig. 2 >>*

143   **2. Stratigraphy of the Hebrides Basin and potential for crustal S contamination**

144   The Mesoproterozoic Torridonian succession of NW Scotland (Fig. 1b) divides into the Sleat, Stoer

145   and Torridon Groups. It is dominated by sandstones and psammites (Stewart, 2002; Kinnaird *et al.*,

146   2007). Minor siltstone and mudrock units are thinner and less extensive than in the overlying

147   Jurassic units. Recent sulfur isotope studies of Torridonian sulphates and sulphides from the far NW

148   of Scotland (Stoer to Gruinard Bay) have identified an isotopic shift in $\delta^{34}S$ associated with bacterial

149   sulphate reduction, highlighting that the Mesoproterozoic terrestrial environment was adequately

150   oxygenated to support life (Parnell *et al.*, 2010; Parnell *et al.*, 2012). For example, pyrite-bearing

151   lacustrine siltstones of the Diabaig Formation (basal Torridonian) have $\delta^{34}S$ compositions of -30.1 ±

152   17.3 ‰. . During the present study, 14 Torridonian, Sleat, and Stoer Group sediment samples were

153   collected from the Isle of Skye and Rum for sulphur isotopic analysis. Of these, only 5 samples

154 yielded enough sulphide precipitate (following whole-rock sulphur extraction – see Section 3.2) to

155 allow for conventional S isotope analysis.

156 The anticipated sulphur concentration of the Lewisian crystalline basement and Mesoproterozoic

157 sediments is significantly lower than in the Mesozoic Hebrides Basin, so while the S-isotopic

158 composition of all of these potential contaminants was analysed during this study, the main focus is

159 laid on the Mesozoic. The sedimentary stratigraphy of the Mesozoic Hebrides Basin is presented in

160 Figure 1b (based on Morton & Hudson, 1995; Hesselbo & Coe, 2000). While several literature

161 sources document the carbon and oxygen isotopic composition of these rocks, only Yallup *et al.*

162 (2013) report the sulphur isotopic composition of a single unit (in the Middle Jurassic Cullaidh Shale

163 Formation; mean bulk rock δ^{34}S -0.7 ± 1.8). Sulphur isotopes for Jurassic sediments from elsewhere

164 in the British Isles have been reported (e.g., Raiswell *et al.*, 1993; Hudson *et al.*, 2001) however until

165 now there has been no Hebridean 'stratigraphy' of sulphur isotopes available in order to establish a

166 S-isotope framework of potential contaminants to BPIP magmas. In the present study, the

167 Palaeogene igneous rocks of interest are observed intruding through, and physically being

168 contaminated by Jurassic sedimentary sequences (via the presence of xenoliths). Thus, it is

169 specifically the Jurassic units which are of most interest for the purposes of this study. This is due to

170 the high proportion and volume of S-rich shales and mudrocks throughout this Jurassic succession,

171 unlike in the older Triassic, or younger Cretaceous sediments that are sulphur poor and sporadically

172 developed across the region (e.g., Hesselbo & Coe, 2000).

173   The Trotternish Sills intrude through the same thick package of Jurassic sediments as the Robustan

174 dykes on the Strathaird Peninsula. Thus equivalent Jurassic sedimentary rocks are present as

175 potential contaminants to ascending BPIP magmas and allow for the two different types of intrusive

176 body (sill vs. dyke) to be tested against one another for their degree of crustal S contamination.

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3. Methodology

3.1 Sampling strategy

In northern Skye, 23 Trotternish Sill Complex samples were collected, including a range of all rock types (picrites, picrodolerites, and crinanites – see Supplementary Tables). Of these, 11 samples were analysed for sulphur isotopes, and PGE and Au.

At Lealt Quarry [NG 5188 6064], a finely crystalline dyke (crystals < 0.5 mm) approximately 2m wide (strike 338°) with a vertical orientation cuts the Trotternish Sill Complex (comprising an upper crinanite sill, overlying a picrite sill and pegmatitic picrite sill – Fig. 3a). The dyke contains 2-20cm centimetre-scale elongate xenoliths of a baked mudrock (likely of Jurassic origin) and 5-10cm picrite sill xenoliths at its margins (Fig. 3b). The dyke appears to be compound with faint vertical banding (showing indistinct boundaries) at its centre, and elongate xenolith trails within some bands (Fig. 3c). This indicates multiple injections of basalt in the dilatational fracture through which the dyke has intruded. Disseminated millimetre-scale (2-10mm) pyrite crystals are associated with these xenoliths and at the dyke margins, with most disseminated pyrite mineralization at the contacts with mudrock xenoliths, or forming 'stringers' through the basalt dyke within 30cm of the xenoliths. There are also trails of white-coloured zeolites (2-10mm diameter) within 20cm of the dyke margin (Fig. 3d). Four samples from the Lealt Quarry basaltic dyke, which cross-cuts the sill units and four Jurassic sediment samples from packages adjacent to or within the Trotternish Sill Complex, were also collected from northern Skye (e.g. Fig. 3e).

In southern Skye, 1 basaltic trachyandesite and 8 basaltic dyke samples were collected from a NW-SE oriented transect approximately 1 km in length through the Jurassic succession at Robustan (Fig. 2). All dykes ranged between 30 cm and 8 m in width, with most measuring 1 – 2.5 m wide. One dyke

202   (SK51) displayed rare quartz-filled amygdales up to 0.5cm in diameter, however all other sampled

203   dykes were amygdale-free. Dyke samples SK124 and SK129 contained visible pyrite within 5cm of

204   the dyke contact with sediments. Samples were collected from a mixture of dyke margins and

205   central zones. In southern Skye, 5 samples of the Jurassic sediments were collected from this area,

206   including shales, a siltstone, and sandstones.

207   *<< insert Fig. 3 >>*

208   *<< insert Table 1 >>*

209

210   **3.2. Whole-rock geochemistry**

211   Major and trace element analyses (including PGE and Au) and S isotopic (δ^{34}S) results are given in

212   Table 1 respectively. A selection of igneous and sedimentary samples were also analysed for

213   elemental S abundance. Sediment S isotopic compositions and S concentrations, spanning the upper

214   portion of the Lower to the Upper Jurassic, are presented in Table 1, and a sub-selection of 4

215   sediments were analysed for their major and trace element geochemistry (Supplementary Material).

216   For the Lower Jurassic, samples were obtained from outcrops on the Ardnamurchan Peninsula,

217   mainland Scotland. In addition, we analysed the S-isotopic composition of 2 sulphide-bearing

218   samples from the Lewisian gneisses of NW Scotland (X66 and X11b), and 1 sample of Moine

219   metapelite from Ardnamurchan (AN78 – Table 1).

220   Unweathered material was crushed, split, and milled to a fine powder in an agate planetary ball mill.

221   Major and trace elements were analysed by inductively coupled plasma optical emission

222   spectrometry (ICP-OES –JY Horiba Ultima-2) and inductively coupled plasma mass spectrometry (ICP-

223   MS - Thermo X Series 2) respectively at Cardiff University using methods and instrumentation

224   described by McDonald & Viljoen (2006). Platinum Group Element (PGE) and Au analysis for samples

225   was carried out by Ni-sulphide fire assay followed by Te co-precipitation and ICP-MS (Huber *et al.*,

226   2000; McDonald & Viljoen, 2006). Accuracy for whole-rock elemental geochemistry was constrained

227   by analysis of the certified reference materials TDB1 and WMG1 for PGE and Au, and JB1a for all

228   other trace and major elements (Supplementary Material). Precision was determined by repeat

229   analysis of a sub-set of samples, with most elements repeatable to within 10% or better.

230   **3.3. Sulphur isotope analyses**

231   Samples with visible sulphide minerals >500μm^2 were cut into blocks (up to 40 x 20 mm in area) and

232   polished. *In-situ* laser combustion of polished sulphides was carried out following the technique of

233   (Wagner *et al.*, 2002). Based on experimental results, laser combustion causes a small and

234   predictable fractionation of sulphur isotope compositions for δ^{34}S of the SO_2 gas produced,

235   compared to the actual δ^{34}S of the sulphide mineral (Wagner *et al.*, 2002). Therefore the raw δ^{34}S

236   data were corrected by the following factor: δ^{34}S$pyrite = \delta^{34}$S$_{SO_2}$laser + 0.8‰. Whole-rock S extracted

237   from the rocks (see below), and a series of samples from which sulphide separates could be picked,

238   were analysed following the technique of Robinson & Kusakabe (1975).

239   SO_2 gas samples were analysed at the Scottish Universities Environment Research Centre (SUERC)

240   using a ThermoFisher Scientific MAT 253 dual inlet mass spectrometer (for conventional samples)

241   and an on-line VG Isotech SIRA II mass spectrometer (for laser combustion samples). Standards used

242   throughout all analyses were IAEA-S-3 and NBS-123 international standards, alongside an SUERC

243   laboratory chalcopyrite standard, CP-1. The results for these gave -31.5‰ (IAEA-S-3, certified to be -

244   31.5‰), -4.5‰ (CP-1, certified -4.6‰) and +17.1‰ (NBS-123; certified +17.1‰), with 2σ < ±0.2‰

245   reproducibility, based on repeated standard analyses. All data are reported in standard per-mille

246   variations from the Vienna Cañon Diablo troilite standard, V-CDT. Full analytical run details can be

247   found in the Supplementary Material.

248   Our Chromium Reducible Sulphur (CRS) sulphide extraction procedure (for whole-rock powders) is

249   based on and adapted from numerous published and unpublished procedures (Zhabina & Volkov,

250 1978; Canfield *et al.*, 1986; Tuttle *et al.*, 1986; Hall *et al.*, 1988; Newton *et al.*, 1995; Nielsen &

251 Hanken, 2002; Labidi *et al.*, 2012). A diagram and written description for the procedure can be found

252 in the Supplementary Material.

3.4. Whole-rock sulphur concentration analysis

254 A selection of powdered rock samples were analysed for bulk rock S concentration by LECO CS230

255 Carbon/Sulphur Determinator at the University of Leicester, UK. Between 0.1 and 1.0 g of sample

256 were used depending on relative bulk S content. Samples were ignited in an O_2 stream and the SO_2

257 produced was analysed by infra-red absorption. Each sample was run in triplicate to monitor

258 precision. Accuracy was monitored by regular analysis of the reference material BAS ECRM 877-1.

259 The limit of minimum detection for this method is 0.018 wt.% S, which is calculated based on 3 x

260 standard deviation of the mean blank value. A total of 11 samples (both BPIP intrusives and

261 sedimentary rocks) were analysed by this method.

262

4. Results of δ^{34}S and S determinations

4.1 Trotternish Sill Complex

265 The δ^{34}S of the Trotternish Sill Complex (including samples from the chilled margins at the base of

266 the sill complex) ranges from +0.1 to -4.9‰, however sample SK102 from 2m above the base of the

267 complex at Skudiburgh, gave -10.8‰ (Fig. 4). Excluding sample SK102, the mean δ^{34}S of the

268 Trotternish Sills is -2.3 ± 1.5‰. Total PGE concentrations for these sills ranges from 12.6 to 30.9 ppb

269 with no clear correlation between whole-rock total PGE content and whole-rock δ^{34}S. Whole-rock Pd

270 concentration ranges from 2.1 to 10.2 ppb and Cu from 57 to 252 ppm (Fig. 5). The Cu/Pd ratio

271 varies from 13300 to 46200 with two anomalously high ratios of 72800 and 81100 (samples SK89

272 and SK83, respectively – SK83 is from the base of the picrite sill complex at Dunflodigarry, δ^{34}S = -

273 4.9‰). Pd/Ir ratio ranges from 3.1 to 14.1 with an anomalous value of 56.5 for sample SK90 (a

274 pegmatitic pircite sill from Lealt Quarry, $\delta^{34}S$ = -2.2‰). Overall, Pt/Pd ratio for the sills varies from

275 0.94 to 2.55.

276 The basaltic dyke at Lealt Quarry, which cross-cuts the Trotternish Sill Complex (Fig. 3a and b), has a

277 $\delta^{34}S$ of -7.5‰ at the margins (Fig. 4a), where the dyke has entrained baked mudrock xenoliths ($\delta^{34}S$

278 signature of -15.4‰; Fig. 6). At the centre of the dyke, where xenoliths are absent, the basalt has

279 $\delta^{34}S$ = -6.5‰. At the dyke margin, the total whole-rock PGE concentration is 17.9 ppb, as opposed to

280 1.18 ppb at the dyke centre. Cu/Pd ratio at the margin is 18650 but is significantly higher in the

281 centre of the dyke (77800). Pt/Pd and Pd/Ir show little variance from the dyke margin to its centre,

282 ranging 12.1 to 14.1, and 1.02 to 1.35 respectively.

283 *<< insert Fig. 4 >>*

284 **4.2 Robustan dyke swarm**

285 The $\delta^{34}S$ of 8 samples of basaltic dykes from Robustan ranges from -2.3 to -30.7‰ (Table 1, Fig. 4a).

286 Sample SK131 (from the centre of a 30cm wide basalt dyke) gave the lightest $\delta^{34}S$, at -30.7‰, and

287 has a measured whole-rock sulphur concentration of 1.339 wt.%. Sample SK129 (from the centre of

288 an 80cm wide basalt dyke) gave 0.408 wt.% S and -15.5‰, and SK127 (from the centre of a 2.5m

289 wide dyke) has 0.122 wt.% S and -5.1‰. This distribution of S isotope composition and S

290 concentration, indicates a strong correlation between the S-isotope signature and concentration of S

291 in the dykes (Fig. 4b) – dykes with a higher concentration of S having significantly lower $\delta^{34}S$

292 (correlation r^2 = 0.98). A basaltic trachyandesite, SK130, with approximately 1 mm diameter rounded

293 pyrite crystallised in some portions of the dyke (mostly at the margins) produced $\delta^{34}S$ of -19.8 ±

294 1.4‰.

295 Total whole-rock PGE concentrations in the dykes range from 6.4 to 52.4 ppb, and Pd ranges from

296 1.25 to 16.2 ppb (Fig. 5a). Cu concentration ranges from 90.3 to 207 ppm (Fig. 5b). Cu/Pd ratios vary

297 between 5590 and 167200. Pd/Ir varies from 11.9 to 48.8 and Pt/Pd ratio ranges from 0.74 to 2.20.

298 The Cu/Pd and Pt/Pd ratios of the dykes are chiefly controlled by Pd concentration (Fig. 5c-d).

299 Transposing all samples onto a NW-SE oriented section (Figs. 2 & 5) we observe that the δ^{34}S

300 composition of the dykes becomes significantly lighter from SE to NW. This broadly coincides with an

301 increase in the stratigraphic level of the Jurassic sediments that the dykes cross-cut, and into which

302 they intrude. Arranging the results in this manner, we see that between samples SK126 and SK131 a

303 sharp decrease in Pd concentration coincides with a considerable shift in δ^{34}S (from -2.9 to -30.7‰

304 respectively) and a sharp rise in Cu/Pd (Fig. 5c). Pt/Pd broadly follows this increasing Cu/Pd trend

305 from SE to NW (Fig. 5d), however SK126 is anomalous due to the significantly higher Pt

306 concentration (25.1 ppb) in this sample.

307 *<< insert Fig. 5 >>*

308 **4.3 Sedimentary and contaminant country rock units**

309 Of the Jurassic sediments collected from Robustan (Fig. 2) and analysed by whole-rock sulphur

310 extraction and conventional analysis, the shale member of the Staffin Shale Formation and the

311 Staffin Bay Formation Carn Mor Sandstone Member have the lightest sulphur isotopic signature,

312 with δ^{34}S = -32.6 and -33.8‰ respectively (Table 1, Fig. 6). The measured sulphur concentration of

313 the shale was 1130 ppm S, but the concentration of sulphur in the sandstone was below detection

314 limit (< 180 ppm). A siltstone member of the Staffin Shale Formation also yielded an average δ^{34}S = -

315 29.2‰. As previously mentioned, the assumed Jurassic mudrock xenolith in the Lealt Quarry basalt

316 dyke, produced δ^{34}S = -15.4‰. Samples of the Staffin Shale Formation (SK103) and Kilmaluag

317 Formation marl (SK108) have very low concentrations of sulphur. Similarly low/absent S contents

318 were recorded in Duntulm Formation limestone. A recrystallized mudstone (Portree Shale

319 Formation) and 2 ironstone samples (Raasay Ironstone Formation with abundant pyrite nodules)

320 from Ardnamurchan produced δ^{34}S = -17.6‰ and -14.7‰ respectively. The pyrite-rich nodules

321 themselves had δ^{34}S = -18.4‰ but had the highest concentration of S with up to 28 wt.% (Table 1).

322   The two pyrite-bearing samples of Lewisian amphibolite gneiss collected from the Assynt Terrane in

323   NW Scotland gave a δ^{34}S of -1.4 to +1.2‰. The Moine metapelite from the Ardnumurchan Peninsula

324   produced δ^{34}S = +2.5‰. Torridon and Sleat Mesoproterozoic sediments collected from the Isles of

325   Skye and Rum (Fig. 6a) have δ^{34}S ranging +1.4 to +4.7‰, although few samples out of the total

326   number collected had sufficient S to produce a useable whole-rock Ag_2S precipitate. Samples from

327   the Aultbea Formation sandstone and a shale from the Diabaig Formation were S poor and did not

328   yield sufficient precipitate for analysis. Together with the Archaean Lewisian gneisses and Moine

329   metasediments, the Mesoproterozoic lithologies delineate a limited range of δ^{34}S, from -1.4 to

330   +4.7‰, with a mean of +2.0 ± 1.8‰. This is clearly distinct from the mean Jurassic sediment δ^{34}S of -

331   21.5 ± 8.0‰ (e.g., Fig. 6) and offers the potential to act as a tracer of upper shallow crustal

332   contamination in the BPIP intrusive suites.

333   << *insert Fig. 6* >>

334

335   **5. Discussion**

336   The results of this study provide the first comprehensive S isotope framework for the BPIP through

337   determining the regional crustal signatures and those of the BPIP magmas on Skye. Our work has

338   identified the following key features:

339   1. Local Precambrian basement has a limited range of δ^{34}S values (-1.4 to +4.7‰) with a mean

340   of +2.0 ± 1.6 ‰, which overlaps typical magmatic values. This contrasts markedly with the

341   Mesoproterozoic values found on the mainland by Parnell et al (2010).

342   2. Local Jurassic sediments have a distinct light δ^{34}S signature ranging -35 to -10 ‰ and a mean

343   of -21.5 ± 8.0‰.

344   3. The flat-lying intrusions of the Trotternish sill complex intruded into Jurassic sediments have

345   restricted δ^{34}S signatures of -5 to 0 ‰ (with one outlier of -10.8‰).

346   4. The late vertical Robustan dykes *cross-cutting the same Jurassic units* have much lighter δ^{34}S

347   ranging from -30 to -2 ‰.

348   Thus the data show that the potential contaminants of the BPIP intrusive rocks on Skye have distinct

349   S isotope signatures, and our study has identified differences in S isotope signature between

350   different styles of intrusion. This provides insights into the sources of S, including local mantle and

351   crustal signatures, and to shed light on models for emplacement and contamination in these

352   intrusives. These aspects are explored in detail below.

353   **5.1. What is the value of the local mantle S isotope signature in the BPIP?**

354   Several studies of the sulphur isotopic composition of oceanic basalts have previously been

355   conducted (Kusakabe *et al.*, 1990; Alt *et al.*, 1993; Labidi *et al.*, 2012). Most recently, the reappraisal

356   of MORB δ^{34}S by Labidi *et al* (2012) indicated that the mantle has a mean value around -1 ± 0.5 ‰.

357   The sulphur content of such basalts is usually coupled with Fe, Ni and Cu, and is either found within

358   sulphides or dissolved in the silicate melt. At magmatic temperatures, fractionation between the

359   dissolved and segregated sulphide fractions of these basalts would be negligible and the bulk system

360   reduced (Ohmoto & Rye, 1979; Labidi *et al.*, 2012). δ^{34}S of MORB and primary plume-derived

361   magmas can thus be anticipated to broadly represent the δ^{34}S of the local mantle, particularly for

362   primitive high-MgO magmatic rocks, where sulphur exsolution is assumed to be minimal, and

363   contamination *en route* from the mantle likely to be limited (e.g., Iceland).

364   S isotope analysis of Icelandic basalts show δ^{34}S from -2.0‰ to +0.4‰, with a mean of -0.8‰

365   (Torssander, 1989). Both tholeiitic and alkaline basalts displayed similar isotopic compositions,

366   although intermediate and acid rocks extend to heavier δ^{34}S, up to +4.2‰. For the Icelandic alkaline

367   and tholeiitic samples of Torssander (1989), the homogeneity of the δ^{34}S distribution provides strong

368   evidence that the sulphur isotopic composition of the parental magmas had not changed

369   significantly from the mantle source to crustal emplacement.

370   The mean δ^{34}S of the Trotternish Sills (excluding one anomalously light sample, SK102) is -2.3‰ ±

371   1.5, which is comparable to the signature of basalts from the Iceland plume (Torssander, 1989) and

372   MORB (Labidi *et al.,* 2012), although the range of sulphur isotopic compositions is somewhat larger.

373   Therefore we have confidence that the picritic Trotternish Sill Complex is representative of the

374   mantle S isotopic signature of the plume in this portion of the NAIP and BPIP.

375   *<< insert Fig. 7 >>*

376   **5.2. Crustal sulphur contamination in dykes from southern Skye**

377   The sulphur isotopic composition and sulphur concentration for the basaltic Robustan dykes (SK127,

378   SK129 and SK131) indicate that the magmas which formed these intrusions were contaminated by

379   crustally-derived sulphur with an isotopically light δ^{34}S signature. Assuming contamination is

380   dominated by the Jurassic rocks a simple binary mixing model may be used, as follows:

381    $\delta^{34}S_{mix} = [(\delta^{34}S_C \cdot X_C \cdot f) + (\delta^{34}S_M \cdot X_M \cdot (1\text{-}f))] / [(X_C \cdot f) + (X_M \cdot (1\text{-}f))]$ (equation 1)

382   where the contaminated magma = $\delta^{34}S_{mix}$; the isotopic composition of the uncontaminated magma

383   and contaminating sediment = $\delta^{34}S_M$ and $\delta^{34}S_C$ respectively; the concentration of sulphur in the

384   uncontaminated magma and contaminating sediment = X_M and X_C respectively; and the fractional

385   abundance of the contaminant mixing with the magma = f.

386   Figure 7 shows a simple binary mixing model for δ^{34}S between an uncontaminated mantle melt and

387   Jurassic sediments (SK50 and SK53). Assuming the δ^{34}S of the contaminant is similar to siltstone

388   SK50, for dyke samples SK126 and SK127, between 2 and 4% input from the sediment S reservoir

389   would account for the measured δ^{34}S in these dykes; whereas for SK129 (δ^{34}S = -15.5‰), 17%

390   contamination is required.

391   For sample SK131, the δ^{34}S of the contaminated dyke is calculable by accounting for 78% of the

392   sulphur isotopic signature as coming from a contaminant (e.g., SK50). Sample SK131 was taken from

393   a 30cm wide basalt dyke. While the field evidence, and whole-rock major and trace element

394   geochemistry indicate that it is likely to be related to the other dyke samples from this area (with the

395   exception of the basalt trachyandesite SK130), the S concentration of this dyke is not representative

396   of the parental magma composition which fed the other Robustan dykes and intruded through

397   neighbouring conduits. Local sulphur concentration could likely be extremely variable in the dykes,

398   particularly if physical entrainment of crustal material is considered. Further, the sulphur

399   concentration within the Jurassic sediments is extremely variable (e.g., sandstone vs. pyrite nodule

400   rich shale). Therefore, as the dykes were intruded through the Jurassic sediment package, they could

401   have experienced contamination by each S-rich horizon in turn as the dyke intruded upwards.

402   Although we cannot eliminate the possibility of multi-component progressive contamination having

403   taken place in the parental magmas of the dykes during ascent, we can use a mass balance

404   calculation to interpret the maximum contamination scenario. Assuming an initial concentration of

405   250 ppm sulphur in the parental basaltic magma (based on primitive mantle concentrations;

406   McDonough & Sun, 1995) for a volume of $1m^3$ of magma in a 1m x 1m x 1m conduit with a 10cm

407   wide baked margin in the wall rocks either side (equivalent to $0.2m^3$ wall rock volume and based on

408   visible field evidence) the concentration of sulphur in the wall rock would need to be approximately

409   7.6 wt.%, assuming a density of $3000kgm^{-3}$ and $2600kgm^{-3}$ for the basalt and shale respectively. This

410   concentration is considerably higher than that measured in SK50 (1130 ppm). Assuming that the

411   entire sedimentary S budget is accommodated within pyrite, this is equivalent to a shale containing

412   14% pyrite. Given that pyrite-nodules are recorded within the Jurassic sediment pile of the Hebrides

413   Basin, with 27.7 wt.% S (e.g., sample AN71; Table 1) this is a feasible composition for the

414   contaminant of SK131. Thus, our model can readily accommodate the observed dyke S

415   concentrations and δ^{34}S if the high variability of S concentration in Jurassic shales and ironstones is

416   taken into consideration. Dyke SK131 could have intruded through an isotopically light, pyrite-rich

417 portion of the sedimentary pile with unusually higher S concentrations than the surrounding shales,

418 thus dramatically reducing the volume of country rock required to be contaminating the magma

419 conduit and accounting for the S-signature of the dyke.

420 **5.3. Contrasting S isotope signatures of sills and dykes on Skye**

421 This study has presented two examples of high-level informs of intrusion from the BPIP and shows

422 that the dykes have incorporated substantial proportions of Jurassic S in both study areas on the

423 Strathaird and Trotternish Peninsulas (e.g., Lealt Quarry). In stark contrast, the sills of the Trotternish

424 Sill Complex display limited sedimentary S contamination, even at the chilled sill margins. Therefore

425 we have a dichotomy – crustally derived sulphur (with a characteristically light isotopic signature) is

426 widely available in high concentrations within the country rocks of both areas, yet other factors

427 appear to control whether or not this is mobilised or preserved in the local magmatic intrusions.

428 The possibility of post-magmatic sulphur mobility must be considered – is the sulphur signature

429 observed in the dykes a feature of contamination during the magmatic event, or has this been

430 imparted post-emplacement via *in situ* bacterial reduction in the basalts themselves, within

431 fractures, or by low-temperature fluid remobilisation? At Strathaird, the dykes are volumetrically

432 small, and surrounded by isotopically light sulphur-bearing sediments. It is plausible that post-

433 emplacement fluids could have circulated through these sediments and through the dykes,

434 imparting the sedimentary δ^{34}S in this manner. However, textural diagnosis for the timing of

435 sulphide mineral formation might be a better indicator. Where present, pyrite forms subhedral

436 crystals up to 2mm in diameter, typically within 20cm of the dyke margin, and less common at dyke

437 centres, although as highlighted above, this has not precluded the light whole-rock δ^{34}S signature

438 from penetrating the dyke centres. As far as can be determined, there are no veinlets or stringers

439 observed connecting these. In hand specimens, there is no correlation between the presence of

440 oxidised fractures and pyrite, and oxidised/weathered material along with any vugs/amygdales was

441 deliberately removed from the sample before sulphur extraction and analysis. Surface

442   contamination from overlying peat bogs is a feature of the geography of both the dyke and sill areas,

443   and therefore is unlikely to be a realistic modern source of S to one, and not the other. Therefore at

444   Strathaird, there is no reasonable evidence to presume that the sulphide signature is not a 'primary'

445   feature.

446   Lastly, we can demonstrate physical evidence of high-level crustal contamination by sedimentary

447   rocks taking place in BPIP magmatic intrusions. The vertical basalt dyke at Lealt Quarry, Trotternish

448   Peninsula, is observed in outcrop intruding through the horizontal picritic Trotternish Sills (Fig. 3a).

449   50-80m below this quarry, the sills overlie the Jurassic Lealt Shale Formation and it is assumed that

450   the vertical basalt dyke extends through the sills and into the sedimentary pile below it. Field

451   evidence of contamination comes from the centimetre-scale mudrock xenoliths entrained and

452   preserved at the dyke margin (Fig. 3b-d). Pyrite crystals (up to several millimetres in diameter)

453   nucleate within the basalt at the xenolith margins and the xenoliths themselves contain minor pyrite

454   (mostly at the xenolith margins). The δ^{34}S of Trotternish Sills at Lealt Quarry are -2.2 and -1.7‰

455   (SK90 and SK92). Critically, this demonstrates that it would not be possible to impart the light

456   sulphur isotopic signature observed in the dyke from sulphur present in the host picritic sills. The

457   mudrock xenoliths, although baked and with no characteristically distinguishing features, are

458   undoubtedly from the Jurassic strata, possibly the Lealt Shale Formation or similar lithology, below

459   the current level of exposure. The δ^{34}S of pyrite within the dyke at its margin is -7.5‰, while the

460   whole-rock sulphur signature of the dyke centre is -6.5‰. The whole-rock sulphur signature of a

461   mudrock xenolith is -15.4‰, suggesting that the light δ^{34}S of the dyke has been imparted from

462   Jurassic mudrocks during dyke intrusion and ascent, and involved their physical attrition and

463   brecciation. Accordingly the intact wall rocks to the dyke were baked and chemically interacted with

464   the intruding magma. This is analogous to the dykes on the Strathaird Peninsula, although xenoliths

465   were not observed here, and is in complete contrast to the Trotternish Sills surrounding this dyke

466   exposure.

467 Mass-independent sulphur diffusion profiles between the Platreef of the Bushveld Complex and

468 footwall country rocks showed that the $\Delta^{33}S$ isotopic ratio is particularly sensitive to wall rock

469 sulphur interaction (Penniston-Dorland *et al.*, 2008). For example, the $\Delta^{33}S$ profile indicates

470 enrichment of S at the Platreef footwall contact via back diffusion controlled by fluids emanating

471 from the cooling and crystallising magma. This process appears to be limited to 5m inside the

472 igneous contact, however the diffusive and advective distances into the country rocks ranges from 6

473 to 9m and 16 to 27m respectively (Penniston-Dorland *et al.*, 2008). In the Lealt Quarry dyke, it is

474 unsurprising therefore, that we observe the margins of the dyke to have a stronger sulphur

475 contamination signature than at the centre, as this is where most magma-sediment interaction and

476 element transfer was taking place during intrusion. Further, we demonstrate that it is highly unlikely

477 that the sulphur isotopic signature of the dyke has been imparted post-emplacement by low-

478 temperature fluids from the immediately surrounding host rocks, as the host rocks are picrite sills

479 with a 'magmatic' $\delta^{34}S$ and are therefore not isotopically light enough to impart the $\delta^{34}S$ found in the

480 basalt dyke.

481 We have established that the strongly negative $\delta^{34}S$ observed in the Skye dykes is a characteristic

482 feature of the magmatic intrusions, inherited from Jurassic sedimentary pyrite with a bacteriogenic

483 $\delta^{34}S$ signature, during the magmatic stage. The question arises, why is this signature so rare or

484 absent in the Trotternish Sills? Is there an intrinsic difference between the sills and dykes and their

485 capacity for local, high-level crustal contamination? After all, the sills and the dykes intrude through

486 the same package of sediments on the Strathaird Peninsula as they do on the Trotternish Peninsula.

487 And if contamination is taking place, is it wholesale melting/contamination (with inputs of all

488 elements within the sediment being mixed with the magma), or is there preferential mobilisation of

489 only the most volatile elements like sulphur?

490 **5.3.1 The role of emplacement mechanism on the crustal contamination signature**

491   During intrusion, many mechanisms can control the upwards movement of magmas through the

492   crust. These include zone melting, penetrative intrusion, and stoping (Oxburgh *et al.*, 1984). The

493   physical method by which a magma body is emplaced could affect the degree of contamination that

494   results. For example, the 'assimilability' of a country rock xenolith entrained in a magma depends on

495   the size of that xenolith, viscous flow (Sachs & Stange, 1993), as well as the mineralogical

496   composition and melting temperature of the xenolith. In addition, the hydration and temperature of

497   the entraining magma will also be a factor. It has been demonstrated that high-Mg basaltic magmas

498   can ascend turbulently if they have a sufficiently high flow rate and conduit width (Campbell, 1985;

499   Huppert & Sparks, 1985) delaying the formation of chilled margins and bringing hot magma in direct

500   contact with country rocks for longer periods of time. Huppert and Sparks (1985) proposed that

501   maximum contamination can occur in dykes which are at the minimum width for fully turbulent flow

502   (approximately 3m) and evidence of turbulent flow in a basalt conduit has previously been described

503   on the Isle of Mull (Kille *et al.*, 1986).

504   The Jurassic mudrocks of the Hebrides Basin will predominantly comprise clay minerals and micas,

505   with variable trace element compositions rich in Al, Si, Fe, Mg and Ti. Early-stage melting of

506   micaceous xenoliths has been shown to occur as an active melt zone at the xenolith rim, depleted in

507   Al_2O_3 and enriched in CaO, MgO and TiO_2 relative to the bulk composition of the xenolith (Shaw,

508   2009). Trace element mobility (e.g., large ion lithophile elements) will also be affected by the water

509   content of this partial melt. We have tried to relate changes in Skye dyke whole rock geochemistry

510   with $\delta^{34}S$ but cannot resolve any correlation between dyke trace element composition and S

511   isotopes. This is possibly because magma compositions were already significantly contaminated by

512   deeper crustal silicic rocks (e.g., Moine and Lewisian basement – see Section 4.4), or due to

513   variations in the original magma compositions (i.e., magma batches) such that trace element

514   variation resulting from later input of Jurassic micaceous material would be masked. In addition,

515   while field relations and hand specimens in our study provide good evidence for mudrock baking, *in*

516   *situ* anatexsis of these crustal rocks is not observed. This is in contrast to millimetre-scale veinlets of

517   anatectic melt a few centimetres from a sill–shale contact (sill is 3 m wide) at Elgol, Skye (Yallup *et*

518   *al.,* 2013).

519   Sulphur and carbon will be mobilised, even without the need for partial melting *senso stricto*

520   (therefore decoupling the trace element geochemistry), and transported as oxidised volatiles

521   released from the baking of the xenolith and/or sedimentary wall rock. Evidence from mudrocks in

522   contact with the Cuillin igneous centre on the Isle of Skye suggests that carbon can remain within

523   these wall-rocks despite severe baking and the high temperatures of the intrusion (Lindgren &

524   Parnell, 2006). However, this Cuillin study did not investigate the preservation of sulphur. The

525   thermal effect of the Cuillin Complex was previously recognised as highly localised, using organic

526   geochemical molecular maturity parameters (Thrasher, 1992). Similarly this localised thermal

527   alteration has been found to be the case in limestone beds underlying the Trotternish Sills at Staffin

528   Bay (Lefort *et al.*, 2012). Nevertheless, sediments underlying sills at Duntulm show a more developed

529   contact metamorphic sequence, including grossular garnet and pyroxene (contact metamorphism)

530   and low-temperature alteration in microfactures containing clays (Kemp *et al.*, 2005).

531   A doleritic sill at Elgol (southern Skye) that intruded through the Middle Jurassic Cullaidh Shale

532   formation demonstrates that both sulphur and carbon may be baked out of sediments in this

533   setting, up to 80 cm from the sill-sediment contact (Yallup *et al.,* 2013). We suggest that a similar

534   amount of sulphur assimilation took place during the intrusion of the Trotternish Sill Complex (scaled

535   to the thicker, and presumably long-lived, sill complex). Yallup *et al.* (2013) also demonstrated that

536     $\delta^{34}S$ became lighter towards the sediment-sill contact, and suggested that this is due to loss of ^{34}S-

537   enriched SO_2 gas during sediment baking. We cannot demonstrate this feature in the Jurassic

538   sediments at the base Trotternish sills due to the extremely low concentration of sulphur remaining

539   in these rocks (almost devoid of S) probably due to substantial baking (and complete loss) of the

540   sedimentary volatile budget.

541   On a global scale, the intrusion of sills (as part of large igneous provinces) into thick organic-rich

542   sedimentary basins has been investigated by Svensen and co-workers. For example, hydrothermal

543   vents originating from the base of sills have been identified in the Karoo Basin of South Africa

544   (Svensen *et al.,* 2006, 2007), Siberian Traps (Svensen *et al.,* 2009) and Vøring Basin offshore Norway

545   (Svensen *et al.,* 2004; Planke *et al.,* 2005). These form when large volumes of volatiles (C, S, H_2O) are

546   baked out of sediments into which magmas have been intruded, and represent significant input of

547   greenhouse gases into the atmosphere at a time coincident with global warming events and/or mass

548   extinctions (e.g., Svensen *et al.,* 2012 and references therein). Most of the aforementioned studies

549   are concerned with the input of C or C-based gases, and have not fully investigated fluxing of S.

550   Nonetheless, in this study we demonstrate the decoupled mobility of S from other trace elements,

551   and highlight the requirement for future investigations into the relative mobilities of C and S (SO_2 is

552   also an important greenhouse gas).

553   As magma ascends between clay-poor and clay-rich horizons, the amount of water and volatiles

554   released will vary, with volatile-rich shales expected to chemically perpetuate the contamination

555   process. Furthermore the mechanical properties of a shale horizon may actively promote magma

556   penetration. At shallow depths, over-pressured shale units can act as 'ductile horizons', permitting

557   sill formation below the expected level of neutral buoyancy, leading to sill inflation and fracturing of

558   the country rock. This has been documented in the NAIP in the Judd Basin where Palaeogene

559   magmas have intruded through a thick sequence of Jurassic-Palaeocene sediments, forming saucer-

560   shaped sills (Thomson & Schofield, 2008). It is possible that a similar mechanism of intrusion was in

561   operation during formation of the Trotternish Sill Complex on Skye (Fig. 8a). We suggest that this

562   entailed a comparatively 'passive' (i.e., predominantly non-brittle) emplacement mechanism for the

563   magma, such that limited brecciation and entrainment of Jurassic country rock material took place.

564   Where sediment-magma interaction did take place, probably at the chilled base of the sill complex,

565   and in the vicinity of within-sill rafts of Jurassic sediments up to 10m in thickness (Gibson & Jones,

566   1991) the sediments became so baked, via contact metamorphism, that volatiles such as S and H_2O

567 appear to have been entirely expelled. Many of the sills formed as a result of multiple injections,

568 therefore magma flowing through the centre of the sill complex (i.e., from later injections) would

569 have been effectively isolated from contamination effects in ~ horizontal the sill conduit itself (see

570 Fig. 8a).

571 The large magma volume represented by the sill complex, and sustained magma throughput (at least

572 in comparison to individual basaltic dykes), could have effectively 'diluted' any crustal sulphur,

573 thereby resulting in a limited change in sulphur concentration and/or $\delta^{34}S$ signature in the magma

574 body. This is potentially analogous to conduit settings in Noril'sk, where sulphides which initially

575 evolved following S-saturation, became flushed out (Brügmann *et al.*, 1993; Lightfoot & Keays,

576 2005). Further, Cu/Pd, S-isotope and S/Se evidence from the sill-like intrusions of the Platreef

577 (Bushveld Complex) suggests that S contamination from crustal rocks occurred early and deeper in

578 the plumbing system below the Platreef (McDonald & Holwell, 2007; McDonald *et al.,* 2009;

579 Sharman *et al.,* 2013). These sulphides were transported away from this deeper zone during

580 renewed/continued magma batch injections and ultimately emplaced in the Platreef itself (Ihlenfeld

581 & Keays, 2011). At Trotternish, prior contamination by deeper crustal material during fractionation is

582 demonstrable through other isotope systems (e.g., Sr, Nd and Pb; Gibson, 1990), and we suggest

583 that contamination continued throughout the ascent of the sill complex's parental magmas.

584 However, it is plausible that the change in the emplacement mechanism, due to the change in

585 orientation (i.e., from vertical ascent to horizontal emplacement) or rheology of the country rocks

586 (i.e., brittle vs. non-brittle deformation), would have significant implications for the conduit shape

587 (Schofield *et al.*, 2012) and degree of magma contamination, and, crucially, influenced element

588 mobility during chemical reaction (Fig. 8a).

589 By contrast, the vertical dykes on Skye show widespread and exceptional degrees of sedimentary

590 sulphur contamination. As previously mentioned, physical brecciation and entrainment of Jurassic

591 mudrocks is observed, and we suggest that the strong crustal chemical 'interaction' of the dykes (as

592 demonstrated by the correlation between sulphur concentration and δ^{34}S; Fig. 4b) is directly related

593 to the magma emplacement mechanism (Fig. 8b). These narrow magma conduits, with

594 comparatively limited magma volume, cooled more rapidly than the sills (as shown by their finer

595 crystalline texture), evidently freezing in the local sulphur contamination signature. Dynamic mixing

596 of magmas between the margins and centre of individual dykes has taken place, thereby imparting

597 the marginal contamination δ^{34}S signature to the dyke centre prior to crystallisation, however this

598 process did not completely equilibrate the crustal sulphur input, so that measurable differences in

599 δ^{34}S of up to 1‰ are observed between margins and centres of a 2m wide dyke. A maximum degree

600 of contamination probably occurred within the initial intrusive pulse of a dyke into any one given

601 sedimentary horizon, with later pulses of magma along that conduit and through the dyke becoming

602 progressively shielded from further contamination. Thus the mechanism of contamination in vertical

603 dykes is likely complicated by multiple injections of magma, forming complex dykes (Fig. 8b).

604 However, most of the examples sampled during this study do not display zoning or banding (with the

605 exception of the dyke at Lealt Quarry), and therefore probably only represent a single magma pulse.

606 *<< insert Fig. 8 >>*

607 **5.4. Crustal sulphur sources and contamination potential for Hebridean Scotland and the BPIP**

608 Identifying the crustal contaminants likely to be responsible for inducing S-saturation in ascending

609 magmas is a key factor in pin-pointing when and where immiscible sulphides and orthomagmatic Ni-

610 Cu-PGE mineralization might have occurred. Therefore we have evaluated the S-isotopic signature of

611 various potential S-bearing contaminants on a more regional scale for the BPIP and Isle of Skye.

612 Aside from the Jurassic sedimentary rocks discussed so far, other upper-crustal sedimentary

613 formations could also be contributors to the sulphur regime. For example, members of the Sleat and

614 Torridon Groups present along the Sleat Peninsula on Skye, and on the Isle of Rum (Fig. 6, Table 1),

615 irrespective of lithology and location, have positive δ^{34}S signatures, ranging +1.4 to +4.7‰. The

616   Kinloch Formation mudstone had the highest δ^{34}S value of +4.7‰ (Fig. 6). While it is possible that

617   BPIP magmas could be contaminated by Mesoproterozoic sediments, it is very unlikely that this

618   material produced the light δ^{34}S signature seen in the sills and particularly the dykes of Skye. This is

619   in contrast to the δ^{34}S results of certain members of the Stoer Group from the far NW Scottish

620   mainland at Stoer Point, where values less than -30 ‰ are recorded (Parnell *et al.*, 2010). If this

621   material is widely present in the crust below the Isle of Skye, it is possible that the sulphur isotopic

622   signatures of some of the BPIP magma bodies presented here could have been contaminated by this

623   Stoer Group material. However Parnell *et al.* (2010) highlighted the patchy nature of this light S-

624   isotope signature, with bacterial sulphate reduction constrained within small continental water

625   bodies, the relicts of which are laterally discontinuous along strike. More directly, there is no surface

626   exposure of the Stoer Group extending as far south as the Isle of Skye – only the equivalent Sleat

627   Group is observed in this BPIP area, and as discussed above, analysis of this material indicates that

628   its S isotope signature is significantly heavier. Therefore the combination of the extremely

629   heterogeneous sulphur isotope signature, and unconstrained subterranean extent of the Stoer

630   Group, and other Torridonian sediments, makes this an unlikely candidate to explain the uniformly

631   light δ^{34}S of the dykes and sills on Skye.

632   There is ample evidence that deeper crustal contamination via assimilation fractional crystallization

633   (AFC) and assimilation during turbulent ascent (ATA), took place across the BPIP. This includes large

634   deep staging chamber settings, or in narrower meter-scale magma conduits. Both Lewisian

635   (granulite and amphibolite-facies) and Moinian contaminants have been successfully shown to

636   explain the Sr, Nd, and Pb isotopic heterogeneities observed in BPIP magmas (e.g., Gibson, 1990;

637   Kerr, 1995; Preston *et al.*, 1998). In particular, Gibson (1990) demonstrated that the Trotternish Sill

638   Complex underwent up to ~20% mid- or lower-crustal contamination, prior to magma injection and

639   emplacement. Thompson (1982) and Gibson (1990) use La/Nb ratios to constrain assimilation of

640   Lewisian rocks by Palaeogene magmas, now preserved as dykes, sills and lavas. There is no

641   correlation between La/Nb ratio and δ^{34}S for dykes included in this study (refer to Supplementary

642 Material). There is a very weak and inconclusive correlation between La/Nb ratio vs. δ^{34}S for sills (r^2 =

643 0.31). Lewisian gneisses have variable, but low sulphur concentrations and δ^{34}S ranging -1.4 to

644 +1.2‰ (this study) and -1.4 to +5.5‰ on the NW Scottish mainland (Lowry *et al.,* 2005). Torridonian

645 sediments measured in this study always have δ^{34}S > 0‰. Moine metapelites also have low or very

646 low concentrations of sulphur, and where measurable have δ^{34}S = +2.5‰ (this study) and +3.4 to

647 +4.6‰ in some psammites (Lowry, 1991). Accordingly, Lowry *et al.,* (2005) found that each crustal

648 terrane across Scotland could be characterised by its dominant crustal units, reflected in the range

649 of δ^{34}S for that region. In the Northern Highland Terrane (which includes Skyes) complexities in the

650 terrane-scale δ^{34}S were suggested as reflecting the thick North Atlantic Craton (i.e., Lewisian and

651 Palaeoproterozoic metasediments of Loch Maree) underlying the upper crustal sedimentary cover.

652 Our study shows that both the Moine and Lewisian deeper crustal materials are unrealistic sources

653 for the consistent and characteristically light sulphur isotopic signature of the Skye dykes. Therefore,

654 we suggest that while deep crustal contamination of BPIP magmas has taken place, the dominant

655 δ^{34}S contamination signature observed in these magmas is the result of specific and localised upper-

656 crustal contamination of readily-fusible, organic-rich Jurassic shales and mudrocks. This has taken

657 place in smaller vertical magma conduits, or where the mudrocks were in direct contact with BPIP

658 magmas, allowing for pervasive alteration, mobilisation of volatiles and in some cases physical and

659 chemical assimilation of these country rocks, modelled by a simple binary mixing scenario.

660 **5.5. Implications for Ni-Cu-PGE orthomagmatic sulphide mineralization**

661 Whilst not the sole forcing factor in the occurrence of S-saturation, significant literature points to

662 the common association of crustal contamination with the development of orthomagmatic sulphide

663 mineralization (e.g., Noril'sk-Talnakh Region (Ripley *et al.*, 2003; Ripley & Li, 2013); Duluth (Arcuri *et*

664 *al.*, 1998); Voisey's Bay (Ripley *et al.*, 1999); Kambalda (Lesher & Burnham, 2001); and others (see

665 Naldrett 2004, 2011 and references therein). A scenario where crustal sulphur is available for

666 magma contamination is therefore often an important factor in Ni-Cu-PGE exploration. A recent

667   study of lavas by Hughes *et al.* (under review) has suggested that Scottish BPIP is one of the most

668   fertile regions of the NAIP for Ni-Cu-PGE mineralisation, in part due to the widespread opportunities

669   for shallow crustal contamination by S-rich sediments.

670   Interpreting sulphide metal tenors and whole-rock chalcophile element abundances of magmas in

671   relation to their degree of crustal sulphur contamination (monitored here by $\delta^{34}S$) can be

672   complicated by the formation of multiple generations of sulphide liquids (i.e., deeper in the

673   magmatic plumbing system; e.g., Holwell *et al.,* 2007) and the effects of prolonged interaction of

674   sulphides with fresh silicate magma(s) (Campbell & Naldrett, 1979). The ultramafic sills have a low

675   Cu/Pd ratio (13300 to 46200) in comparison to most of the basaltic Robustan dykes (5590 and

676   167200), however this ratio is still elevated above that of primitive mantle (c. 7000-8000 based on

677   pyrolite; McDonough & Sun, 1995), suggesting that sulphide saturation had probably already taken

678   place prior to emplacement of the sills. The highly variable Cu/Pd ratio of the Robustan dykes is

679   likely also recording one or more S-saturation events which occurred prior to dyke emplacement.

680   However because the dykes are demonstrably assimilating local crustal sulphur at the current

681   erosion level, we can further inspect the response of the remaining low-abundance chalcophile

682   elements to this secondary S-saturation event.

683   Broadly, dyke samples with the lightest crustal $\delta^{34}S$ signatures have the lowest Pd concentration (Fig.

684   5a) and highest Cu/Pd ratio (Fig. 5c). Pt, Pd and Au concentrations show the strongest response to

685   localised crustal S contamination, and variations in Cu appear to be weaker (i.e., less variable

686   concentrations) in this secondary S-saturation scenario. This causes a correspondingly 'delayed'

687   spike in Cu/Pd ratio. In PGE mineralised settings (e.g., the Bushveld Complex) this feature is well

688   documented for S-saturation in PGE reefs (Maier, 2005 and references therein) as the partition

689   coefficient of Pd into sulphide liquid is several orders of magnitude greater than that for Cu (Mungall

690   *et al.,* 2005; Mungall & Brenan, 2014). However in a setting where first-stage S-saturation has

691   already occurred deeper in the system, the concentration of chalcophile elements (particularly Pd-

692   group PGE) would be much lower in the remaining silicate magma that continued its ascent to the

693   surface. Therefore the Cu/Pd ratio of that residual silicate magma would be much higher and more

694   variable than that of a magma which had only undergone one S-saturation event (as seen in the

695   Trotternish Sills). From this secondary 'starting point' Cu/Pd ratio, a second-stage localised S-

696   saturation event would lead to further fractionation of Pd from Cu (particularly for systems with low

697   R-factors; the ratio of sulphide liquid to silicate magma), raising Cu/Pd ratio to even higher values

698   (up to 167,000 in sample SK131). Assuming sulphide liquids from the initial S-saturation event had

699   not been entrained during continued magma ascent, the extreme variability in Cu/Pd ratio caused by

700   this first S-saturation (deeper) would lead to even greater erraticism in this parameter during the

701   formation of later sulphide liquids (shallower). Thus, in the absence of entrained sulphide liquids, the

702   detailed interpretation of chalcophile element ratios at/after secondary S-saturation events would

703   be impractical.

704   The cause for this first-stage S-saturation event remains speculative – given the silicic basement

705   underlying this region, this could have been due to the addition of SiO_2 to the magma (as well as

706   fractional crystallisation during magma ascent) such that S concentration at sulphide-saturation

707   (SCSS) was lowered. Alternatively this could have been triggered by addition of S deeper in the crust,

708   but during this study we have shown that $\delta^{34}S$ of Moine, Torridonian and Lewisian rocks is within

709   magmatic variation (0 ± 4‰) and therefore would not be detected by this method. However, we

710   have also determined that S abundance in these lithologies is typically very low, and we question the

711   likelihood of these rocks in contributing enough S during deep crustal contamination to induce S-

712   saturation.

713   Ultimately the effects of multiple S-saturation events might be further investigated by analysis of the

714   S/Se ratio of sulphide minerals (e.g., Ihlenfeld & Keays, 2011). In addition, the earlier S-saturation of

715   magmas would likely preclude significant orthomagmatic Ni-Cu-PGE mineralization in higher

716   portions of a magmatic province. However, by identifying the physical and structural factors

717   controlling the ability of an ascending magma to assimilate crustal sulphur in a conduit, we can

718   ascertain the locations within a magmatic plumbing system (deep or shallow) where S-saturation is

719   likely to have taken place, leading to mineralization.

720   Finally, the small (± 1.4‰) and non-systematic variation observed between δ^{34}S of sulphide minerals

721   (in this case, pyrite) compared to whole-rock values is acceptable for use in identifying the

722   occurrence of contamination of magmas by crustally-derived sulphur. The observed maximum

723   procedural-induced isotopic shift is very small in comparison to the isotopic shift caused by crustal

724   contamination and the natural δ^{34}S of the country rock samples. However this is dependent on the

725   crustal sulphur source having a characteristic δ^{34}S signature (either light or heavy), and being

726   statistically different from the typical 'mantle' signature. Therefore the whole-rock CRS method of

727   sample preparation for δ^{34}S analysis (as used in this study) is suitable for identifying areas of

728   potential sulphur saturation in mantle-derived magmas contaminated by crustal sulphur. This

729   contributes to a rapid and flexible exploration tool for orthomagmatic Ni-Cu-PGE sulphide deposits,

730   applicable even to samples with non-visible sulphide mineralization, as it identifies crustal sulphur

731   contamination of whole-rock magmatic samples, assuming that there is a significant difference

732   between the sedimentary and mantle δ^{34}S composition. This method offers the opportunity to

733   vector into areas of increased crustal S contamination, even in relatively S-poor rocks.

734

735

6. Conclusions

736

737   1. The δ^{34}S range for the Palaeogene alkali picritic Trotternish Sill Complex (+0.1 to -4.9‰; mean

738   = -2.3 ± 1.5‰) reflects the dominance of mantle-derived sulphur, consistent with that found

739   in other Icelandic mantle plume-derived magmas.

740   2. In contrast, the observed δ^{34}S range for the Palaeogene basaltic Robustan dykes is -2.3 to -

741   30.7‰. Sulphur isotope evidence for contamination is present throughout the full widths of

742   these dykes: margins and centres alike.

743   3. There appears to be a fundamental geological control on the degree of crustal contamination

744   of magma bodies, depending of the mechanism (orientation and energy) by which that

745   magma intruded. In spite of similar country rocks characterised by light δ^{34}S (> -33.8‰), there

746   is a distinct difference in the S contamination pattern between a flat-lying concordant and

747   thicker magma conduit (as represented by the 10-100m-scale Trotternish Sill Complex, δ^{34}S

748   from -4.9 to +0.1‰ with one outlier of -10.8‰), and a vertically intruded discordant and

749   narrow magma conduit (meter-scale basaltic Robustan dykes, δ^{34}S from -2.3 to -30.7‰).

750   4. Turbulent flow within narrow magma conduits emplaced discordantly to sediments maximises

751   contamination potential, particularly of volatile and highly mobile elements (such as S) in a

752   high surface-to-volume scenario. Whereas passively emplaced, wide conduits emplaced

753   concordantly to the sediments undergo comparatively little crustal sulphur contamination.

754   This mechanical control may act in addition to other known controls on contamination such as

755   the temperature and flux rate of magma in a conduit system.

756   5. Despite the absence of sulphide minerals, a S-saturation event prior to dyke emplacement

757   may be identified using Cu/Pd ratio. Given the high concentration of S in these dykes, and

758   their strong δ^{34}S crustal contamination signature, S-saturation causing this extreme variability

759   in Cu/Pd ratio must have been imparted before dyke emplacement and therefore at a deeper

760   crustal level. This suggests that Ni-Cu-PGE mineralisation may exist below current exposure

761   levels. However the trigger for this previous S-saturation remains unresolved.

762   6. Where sulphide minerals are either too rare or too small to separate, use of whole-rock

763   sulphur extraction to analyse δ^{34}S in samples can reveal S contamination. We suggest that this

764   technique offers a suitable and rapid method that can be used as a vectoring tool for

765   exploration of orthomagmatic Ni-Cu-PGE mineralization.

766

775

Figure 1 – (a) Location and simplified geological map of part of the British Palaeogene Igneous Complex (BPIP) – the geology of the Isle of Skye, Rum and the Small Isles. Two black boxes highlight close-up geological maps (see Supplementary Material B). Light grey background does not delineate specific geology, but we highlight that the Lewisian gneisses underlie this entire region (north of the Great Glen Fault). (b) Hebrides Basin sediments area surrounding the Isle of Skye and the Small Isles. Adapted from Fyfe *et al.,* (1993).

Figure 2 – Strathaird Peninsula (southern Skye). Line of cross-section displays position of the dyke samples as projected parallel to the strike of the sediment bedding planes, with the exception of sample SK40, which has been projected onto the cross-section along the strike of the dyke itself. Dykes drawn assuming vertical dip. Sediments dip approximately 10° dip NNW below the sediment-lava contact at Scaladal Burn (note cross-section shows 2 x vertical exaggeration). Extent of dyke structure under the current surface level is unknown – cross-section projects dykes for display purposes only.

Figure 3 *(colour online)* **–** Annotated photographs of field relations for the Trotternish Sills and cross-cutting dykes on the Trotternish Peninsula. (a) Lealt Quarry (looking West) – Horizontal Trotternish Sills cross-cut by a later, banded basalt dyke. (b) Close-up of Northern margin of basalt dyke in Lealt Quarry showing both picrite (white lines) and mudrock (black lines) xenoliths in a fluid-rich coarser section of the dyke approximately 10cm from the chilled margin. Some mudrock xenoliths have begun to disintegrate and 'fray' at the edges, becoming assimilated or with ribbon-like fluid-rich trails extending into the basalt. (c) Faint banding in the central portions of the dyke. Some 'bands' have elongate mudrock xenolith trails. (d) Close-up of the Southern margin of the basalt dyke at Lealt Quarry showing trails of white zeolites close to the chilled margin (annotated by black dashed

799   circles). Baked mudrock xenolith with white coloured fluid-rich stringers extending into basalt shown

800   at top of the image. Basalt dyke contact with picrite sill is annotated by solid white line, and c. 5mm

801   wide basalt chilled margin by dashed white line. (e) Skudiburgh, looking North, with vertical basalt

802   dyke cross-cutting the horizontally banded Trotternish Sills. No offsets of dyke or sills, despite the

803   angle of the photograph. Base of the sill package delineated by black solid line, which overlies a

804   poorly exposed area of baked mudrocks (best observed in the area delineated by dashed black

805   lines).

806   **Figure 4 –** Sulphur isotope (δ^{34}S) results for Isle of Skye sills and dykes (a) All sills and dykes. (b)

807   Correlation between whole-rock S concentration and δ^{34}S for Robustan dykes (line of best fit is a

808   logarithmic fit as shown, but a linear fit also produces r^2 value > 0.95). See main text in Section 4.2

809   for further details.

810   **Figure 5 –** Sulphur isotope (δ^{34}S) results (grey histograms) and whole-rock PGE and Cu

811   concentrations and ratios (black lines with circles) for the basaltic Robustan dykes, aligned according

812   to a NW-SE transect, as displayed in the cross-section of Figure 2. (a) Pd (ppb) and δ^{34}S, (b) Cu (ppm)

813   and δ^{34}S, (c) Cu/Pd x 1000 ratio and δ^{34}S, and (d) Pt/Pd ratio and δ^{34}S. The light grey shaded areas in

814   (c) delineates the range of Cu/Pd ratios measured in samples from the Trotternish Sill Complex and

815   dark grey delineates mantle ranges (according to McDonough & Sun, 1995).

816   **Figure 6 –** Sulphur isotope (δ^{34}S) results for Jurassic and Torridonian (Mesoproterozoic) sediment

817   samples from the Hebrides Basin area, Isle of Skye, Isle of Rum, and Ardnamurchan.

818   **Figure 7 –** (a) Simple binary mixing model between a mantle melt (starting δ^{34}S = 0.0‰, 250ppm S)

819   end member, and a contaminating end member of either Staffin Bay Formation sandstone, sample

820   SK53 (δ^{34}S = -33.8‰, < 180ppm S) or Staffin Shale Formation shale unit, sample SK50 (δ^{34}S = -

821   32.6‰, 1130 ppm S). Overlain on the plot are basalt dyke samples SK127, SK128, SK129 and SK131,

822   with estimates of 'f' (mass fraction of S from the contaminating end member) for each sample.

823 **Figure 8 –** (a) Passive intrusion of magmas forming sills – while the ~ vertical oriented feeder zone(s)

824 are likely to actively assimilate and become contaminated by wall rocks during magma ascent, once

825 the magma reaches a ductile horizon or zone of neutral buoyancy (spreading out horizontally) it is

826 emplaced in a 'passive' environment of inflation. In this 'passive' setting, little physical attrition of

827 wall rocks will occur so that little/no crustal S-isotope signature is imparted. (b) Active intrusion of

828 magmas forming dykes – the ~ vertical and high-energy intrusion of magmas into upper crustal levels

829 forming dykes (e.g., at Lealt Quarry or Robustan) causes physical attrition/brecciation of wall rocks.

830 This higher energy (or 'active') or mechanism of emplacement leads to the entrainment of xenoliths

831 (via wall rock attrition) and chemically leaches wall rocks of volatiles (S), imparting a crustal S-

832 isotopic signature. In both cases (a) and (b), the contamination signature is inferred to be a local

833 effect (magmas have transported the signature on a maximum scale of 100's meters). See main text

834 (Section 5.3.1) for further details.

835

836

837 **Table captions**

838 **Table 1 –** Summary of sulphur isotopic compositions ($\delta^{34}S$) measured by conventional methodology

839 (following whole-rock sulphur extraction; indicated as 'wr') and laser combustion methodology

840 (indicated as 'Laser'). Whole-rock S concentration (in wt. %) is also listed. In instances where a

841 powdered sample was subjected to the whole-rock sulphur extraction (CRS) method, but no

842 precipitate was yielded or the precipitate was too small to facilitate conventional analysis, 'no

843 precipitate' is annotated in Table 1. (*)Indicates that the S concentration of this sample was too high

844 to be determined (based on the calibrated set-up of LECO) and thus the estimated S concentration is

845 listed in Table 1. Note that samples with S concentration below detection limit are indicated by <

846 0.02 (detection limit). Full mineral-specific and duplicate sample details are available in the

847 Supplementary Material.

848

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Fig. 1

Fig. 2

Cross-section scales:
Strathaird section A-B: 2 x vertical exaggeration

NW
Beinn Leacach
SK40
SK51 SK50
SK52
SK131
SK130
SK129
SK127, SK128
SK126
SK125
SK124
Robostan
SE

Mesozoic sediments:

Upper Cretaceous sandstones and conglomerates

Upper Cretaceous Strathaird Limestone Formation

Staffin Shale Formation (shale)

Staffin Bay Formation (sandstone)

Skudiburgh Formation (siltstone)

Kilmaluag Formation (calcareous mudstone)

Duntulm Formation (mudstones and limestones)

Valtos Sandstone Formation (muds and limestones)

Lealt Shale Formation (muds, sands and limestones)

Elgol Sandstone Formation (sandstone)

Cullaidh Shale Formation (mudstone)

Bearrreraig Sandstone Formation (sandstone)

Raasay Ironstone Formation & Portree Mudstone Fm

Scalpay Sandstone Formation (sandstones and silts)

Pabay Shale Formation (mudstones)

Cross-secton legend:

Picrobasalt

Basalt

Hawaiite

Mugearite

SK51 Dyke sample

SK50 Sediment sample

0   1000
             m

Fig. 3

a)

Crinanite

Picrite

Picritic pegmatite

Picrite

Trotternish Sills
(~ horizontal)

Rubble and
loose material

Compound dyke?
(subtle banding
between chilled
margin and central
sections)

Basalt dyke
(cross-cutting
Trotternish Sills)

Inset for photo (b)
Inset for photo (c)

3 m

b)

Baked mudrock
xenoliths

Ribbon-like
assimilation
of mudrock
into basalt dyke

Fluid-rich
'pockets' at
dyke margins
within 20-30 cm
of true chilled
margin

10 cm

Coarse picrite xenoliths

c)

Compound basaltic dyke
(dashed lines inferring
multi injections of magma,
boundaries sometimes indistinct)

Dyke chilled margin (N side)

Trails of thin/elongate
baked mudrock xenoliths
entrained within some
central basalt dyke bands

20 cm

d)

Picrite sill
(~ horizontal)

Mudrock
xenolith
(fluid-rich
veinlets
feeding
into basalt)

Basalt dyke
(~ vertical)

Sill-dyke
contact

Zeolite
trails

Dyke chilled
margin
(~ 5mm wide)

1 cm

e)

Sill-sediment contact
(base of sill complex?)

Banding and
layering
in sills

Exposure of baked fissile
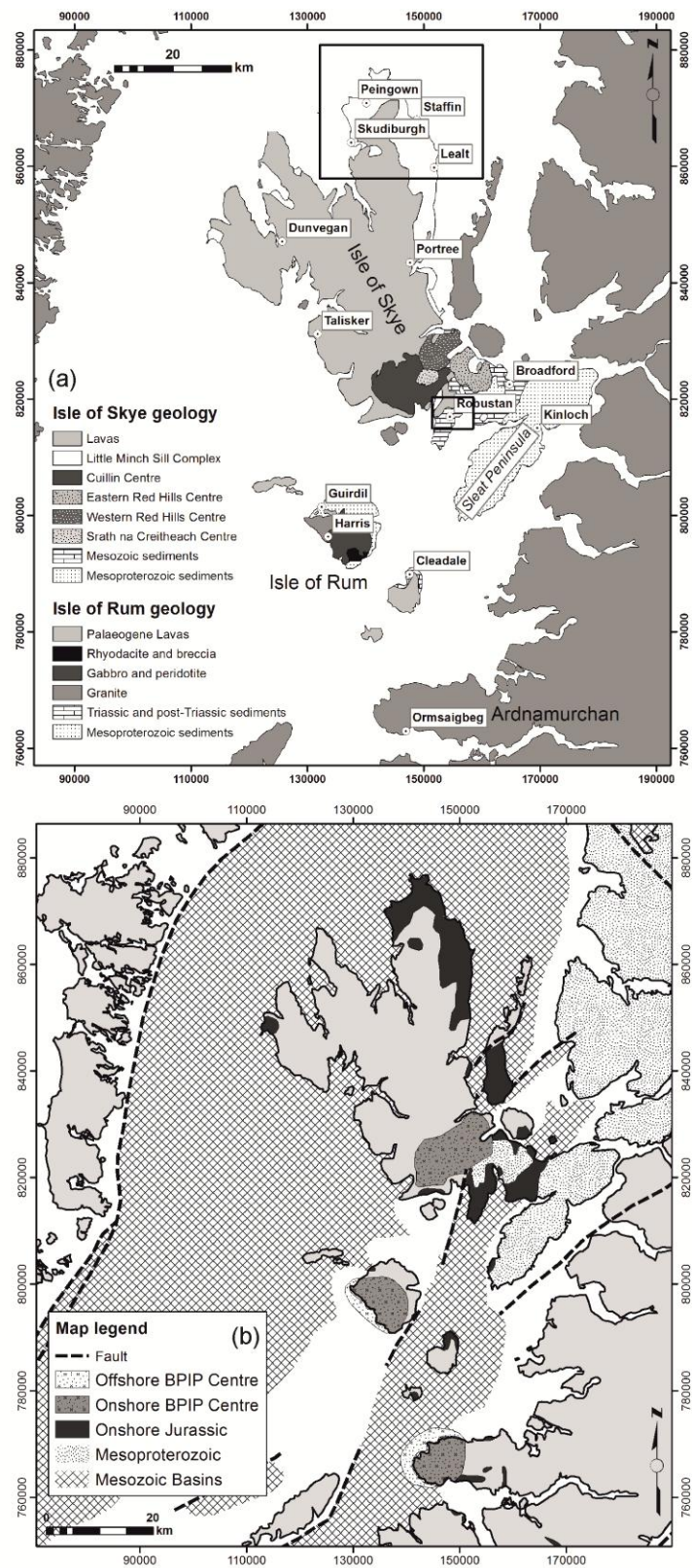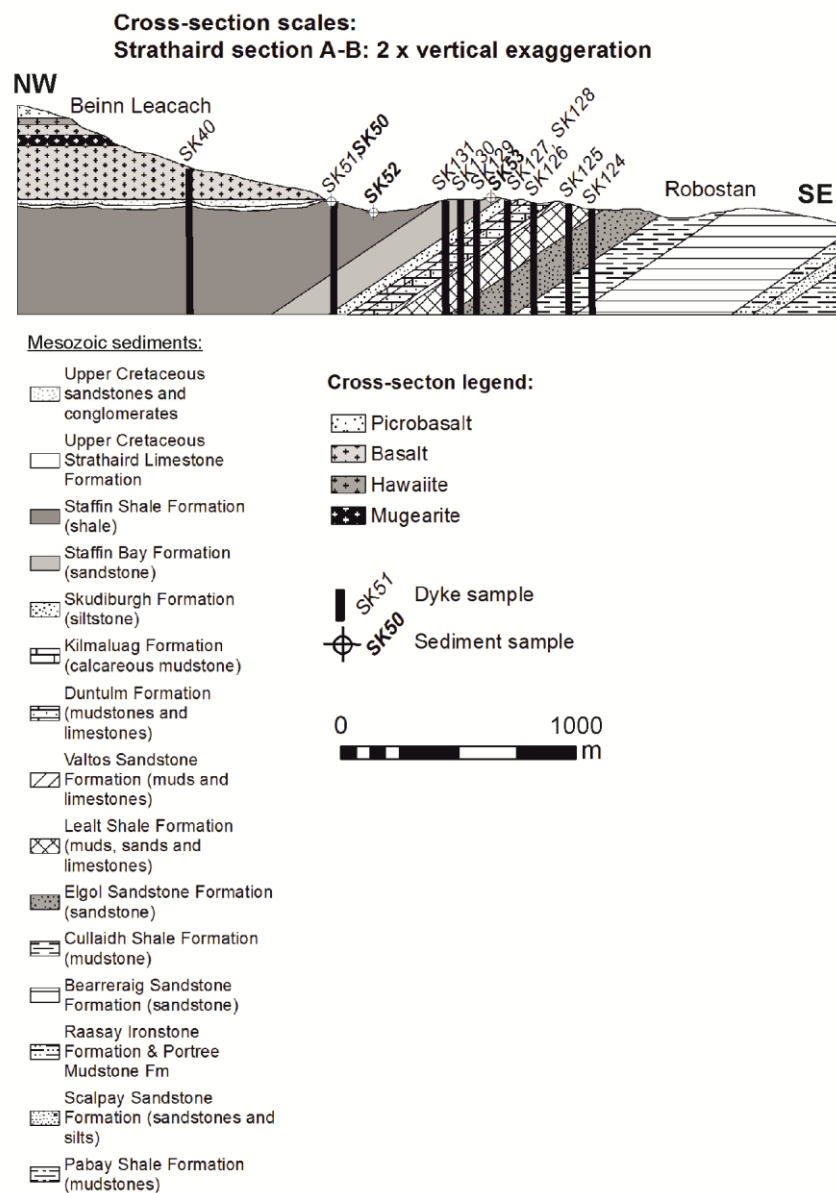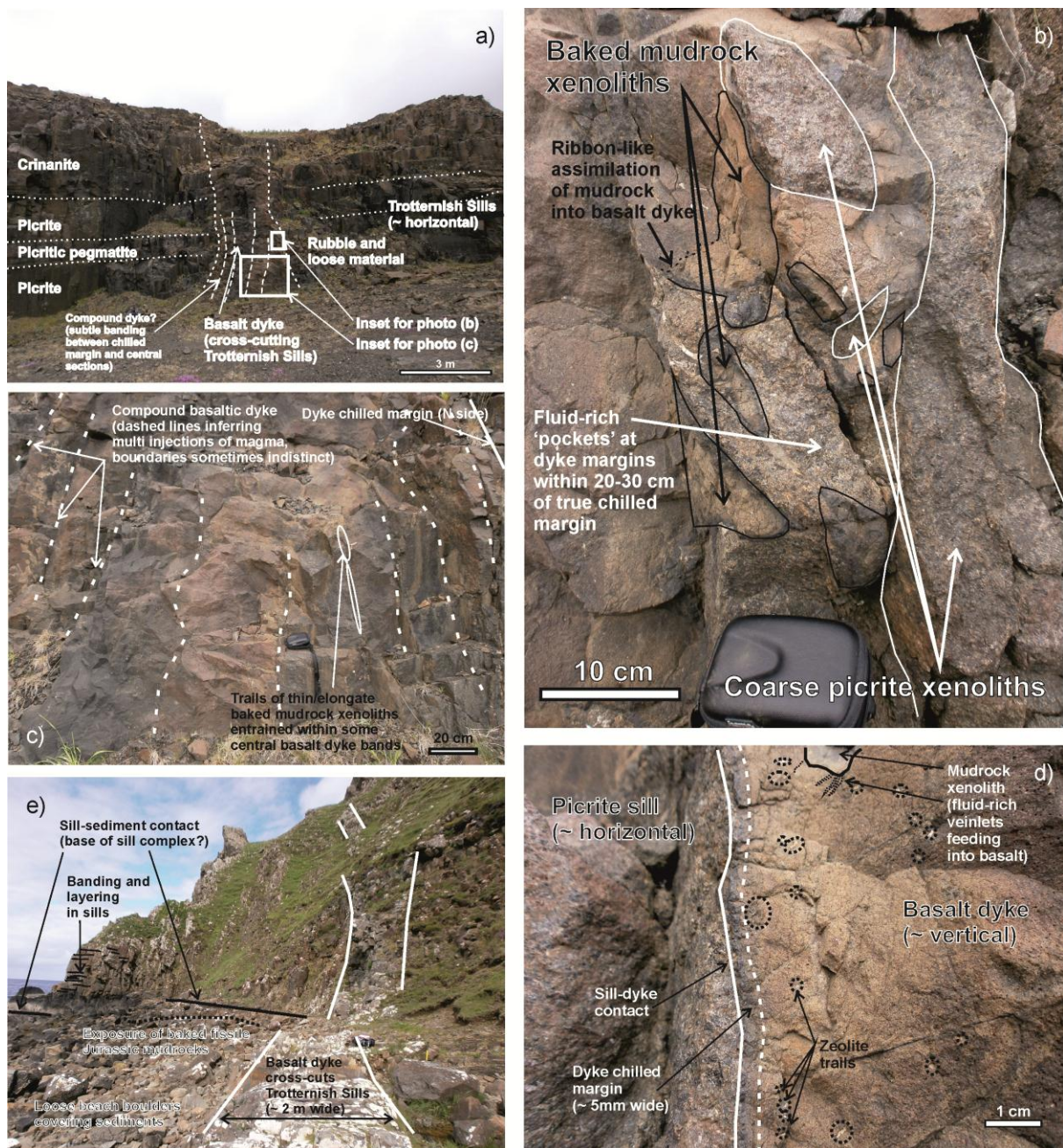Jurassic mudrocks

Loose beach boulders
covering sediments

Basalt dyke
cross-cuts
Trotternish Sills
(~ 2 m wide)

Fig. 4

Fig. 5

Fig. 6

Fig. 7

Fig. 8

a) 'Passive' intrusion for sills

'Passive' sill propagation by inflation of multiple magma injections (complex sill) along a neutral buoyancy horizon

Change in orientation of magma intrusion between ~vertical feeder to ~horizontal sill corresponds to change in energy (drop-out of entrained xenoliths)

Thick package of Jurassic mudrocks

Present-day exposures of the Trotternish Sill Complex?

'Active' wall-rock assimilation

Physical brecciation & entrainment

Feeder zone

b) 'Active' intrusion for dykes

'Active' wall-rock assimilation

Thick package of Jurassic mudrocks

Physical brecciation & entrainment of Jurassic mudrocks

Banded dykes with continued dilation of fractures

Present-day exposures of basalt dykes at Robustan?

Upper crustal ascent and injection of magmas