Journal of Archaeological Science: Reports 58 (2024) 104738

Contents lists available at ScienceDirect

Journal of Archaeological Science: Reports

journal homepage: www.elsevier.com/locate/jasrep

Was the Stonehenge Altar Stone from Orkney? Investigating the mineralogy and geochemistry of Orcadian Old Red sandstones and Neolithic circle monuments

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ARTICLE INFO

Keywords:
Altar Stone
Stonehenge
Orkney
Old Red Sandstone
Sandstone
Provenancing

ABSTRACT

Recent petrological, mineralogical and geochemical investigations of the Stonehenge Altar Stone have negated its source in the Old Red Sandstone (ORS) Anglo-Welsh Basin. Further, it has been suggested that it is time to look wider, across northern Britain and Scotland, especially in areas where geological and geochemical evidence concur, and there is evidence of Neolithic communities and their monuments. In this context the islands of Orkney, with its rich Neolithic archaeology, are an obvious area worthy of investigation. The same techniques applied to investigations of the Altar Stone and ORS sequences in southern Britain have been applied to two major Neolithic monuments on Mainland Orkney, namely the Stones of Stenness and the Ring of Brodgar. In addition, field samples of ORS lithologies from the main stratigraphic horizons on Mainland Orkney have been investigated.

Portable XRF analyses of the five exposed stones at the Stones of Stenness and seven of the exposed stones at the Ring of Brodgar show a wide range of compositions, having similar compositions to field samples analysed from both the Lower and Upper Stromness Flagstone formations, with the stones at Stenness appearing to have been sourced from the Upper Stromness Flagstone Formation while the Ring of Brodgar stones possibly being sourced from both formations. Examination of the mineralogy of ORS field samples and the Stonehenge Altar Stone, using a combination of X-ray diffraction, microscopy, Raman spectroscopy and automated SEM-EDS shows there to be no match between the Orkney samples and the Altar Stone. Only two samples from Orkney showed the presence of baryte, a characteristic mineral of the Altar Stone. Another key discriminant is the presence of abundant detrital K-feldspar in all of the Orkney field samples, a mineral which has only very low abundance in the Altar Stone. In addition, the regularly interstratified dioctahedral/dioctahedral smectite mineral tosudite is present in the clay mineral assemblage of the Altar Stone, but not detected in the Orkney samples.

It is concluded that the Altar Stone was not sourced from Mainland Orkney, despite considerable evidence for long-distance communications between Orkney and Stonehenge around 3000/2900 BCE.

1. Introduction

Thomas (1923) provided the first detailed account of the Stonehenge bluestones, 'foreign stones' exotic to the Wiltshire landscape in which they currently sit. He proposed that the igneous component in the bluestone assemblage came from the Mynydd Preseli area in north

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https://doi.org/10.1016/j.jasrep.2024.104738
Received 23 July 2024; Accepted 22 August 2024
Available online 30 August 2024

Pembrokeshire (Fig. 1), southwest Wales, identifying sources from Ordovician dolerite and rhyolite outcrops at Carn Meini, Cerrigmarchogion, Foel Drygarn and Carn Alw. In contrast, the Altar Stone is a grey-green sandstone which is anomalous within the rest of the bluestone assemblage in terms of its lithology, size, weight and age. It has long been assumed to be Devonian in age (see Maskelyne, 1878; Thomas, 1923), belonging to the Old Red Sandstone (ORS) Supergroup, and has a mineralogy characterised by the presence of calcite and baryte cements, with the regularly interstratified dioctahedral chlorite/dioctahedral smectite mineral tosudite prominent in its clay mineral assemblage, and only trace amounts of detrital K-feldspar. The Altar Stone also occupies an anomalous position in the Stonehenge landscape, lying towards the centre of the monument, away from the bluestone circle and bluestone horseshoe. In addition, there is no constraint on when the Altar Stone arrived at Stonehenge. The majority of the bluestones are thought to have arrived during construction Stage 1 in the Late Neolithic, *c.* 2900 BCE (Parker Pearson, 2023) and to have been placed initially in a set of holes known as the Aubrey Holes. However, there is no evidence for the Altar Stone having been part of that Stage 1 construction event. It is even not known whether the Altar Stone ever stood upright.

Thomas (1923) considered that the Altar Stone came from either the ORS Cosheston Group near Milford Haven in south Pembrokeshire (Fig. 1), or alternatively from the ORS Senni Beds exposed to the east across the historic county of 'Glamorganshire' (comprising parts of the current counties of Carmarthenshire, Powys and Monmouthshire). However, recent studies by Bevins et al. (2022a, 2023a) concluded that the Stonehenge Altar Stone (Stone 80) is unlikely to have been sourced in Wales or the Welsh Borderland (from the so-called Anglo-Welsh ORS Basin) and have argued that investigations needed to 'broaden horizons', both geographically and stratigraphically, going as far as suggesting that the Altar Stone should not be included within the bluestones. Bevins et al. (2023a) suggested that possible sources for the Altar Stone should have affinities with continental "red-bed" sedimentary rocks with elevated-Ba concentrations and also with known

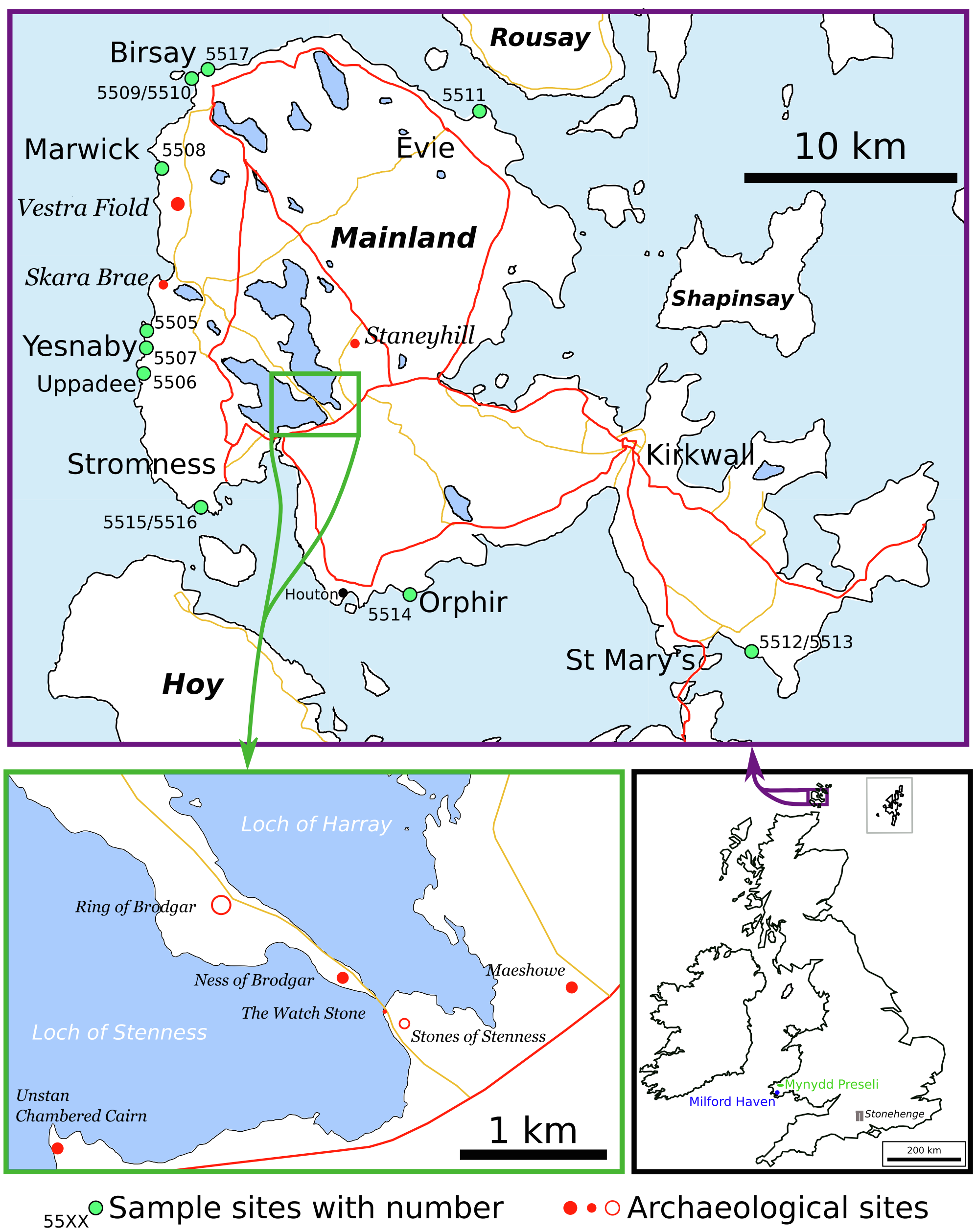

Fig. 1. Map of Mainland, Orkney and its surroundings. Locations mentioned in the text are indicated on the map. Inset shows the area around the Ness of Brodgar where the Ring of Brodgar and Stones of Stenness are located. Also shown are the Old Red Sandstone field sampling sites with sample numbers.

Neolithic activity.

Prompted by the need to start searching elsewhere for a source for the Altar Stone, age dating of zircon, rutile and apatite grains from the Altar Stone has been undertaken to constrain possible source sedimentary basins. Results compared with published age dates for those minerals in sandstones from ORS basin sequences in Britain led to the spectacular conclusion that the Altar Stone was most probably sourced from the Orcadian Basin sequences of mainland and offshore NE Scotland, at least 700 km from Stonehenge (Clarke et al., 2024). An obvious potential source lies in the Orkney Islands, given its extensive array of Neolithic structures, including the Ring of Brodgar, the Stones of Stenness, Maes Howe, Skara Brae and the massive complex of monumental Neolithic buildings at the Ness of Brodgar. Significantly, there is abundant evidence for long distance connections between Stonehenge and Orkney around 3000/2900 BCE, which includes Orcadian-style Grooved Ware pottery being found in pits at Bulford, on the east side of the River Avon, *c.* 4 km east of Stonehenge (Leivers, 2021), as well as a macehead made from Lewisian Gneiss found in a burial place close to Aubrey Holes 18 and 19. Parker Pearson (2023, 44) considers that this was a grave good and although the stone was probably sourced in the Outer Hebrides, it was most probably manufactured on Orkney where similar maceheads are known to have been made (Anderson-Whymark et al., 2017). Some 500 years later, during the second stage of construction at Durrington Walls, *c.* 3.2 km northeast of Stonehenge, around 2500 BCE, hundreds of dwellings were built most probably to house the monument's constructors; these dwellings have the same size and layout as those at Skara Brae, on Orkney (Parker Pearson 2023, p. 60). Bradley (2024) has recently highlighted evidence for long distant links between a number of prehistoric sites in Late Neolithic Britain and Ireland which share architectural elements, suggestive of social connections across vast distances, in some cases up to 400 km. However, this suggestion of a 'unified culture across Britain – pottery, house plans, burial practices and henges' both in Orkney and at Stonehenge is disputed by Barclay and Brophy (2020) who stress the uniqueness of the Orkneys and suggest that there was a non-unified cultural variation within the late Neolithic of Britain.

As mentioned earlier, the Altar Stone contains the mineral baryte and as a consequence shows high Ba contents. Therefore, sandstone sequences with high Ba contents are a logical target to identify sampling areas for comparison with the Altar Stone. The Orkney islands are predominantly composed of sandstones, siltstones and mudstones of Middle ORS (Mid Devonian) age. Interestingly, the British Geological Survey stream sediment map (Everett et al., 2019) shows elevated Ba concentrations in the area around Yesnaby, in SW Mainland (Orkney) in Lower ORS sandstones (see Bevins et al., 2023a), in close proximity to the most significant Neolithic remains.

This paper presents the findings of an investigation into the geochemistry of the monoliths comprising the Stones of Stenness and the Ring of Brodgar stone circles on Orkney and compares the findings with results obtained from the Altar Stone (and derived fragments) presented by Bevins et al. (2020, 2022a, 2023b) in order to ascertain whether or not there is any correlation between the Orkney stones and the Stonehenge Altar Stone. In addition, we present mineralogical and geochemical data for ORS outcrop samples from Mainland Orkney and explore the potential for using these kinds of data to trace orthostats from the Ring of Brodgar and the Stones of Stennes back to their source sites of extraction. Fig. 1 shows the distribution of the main Neolithic monuments on Mainland Orkney, along with the locations of the ORS samples investigated as part of this study.

2. The investigated stones

2.1. Stonehenge Altar Stone

The Stonehenge Altar Stone (Stone 80) has been intensively investigated recently using a range of techniques, including transmitted and reflected light microscopy, portable XRF (pXRF), automated scanning electron microscopy (SEM-EDS), Raman spectroscopy and X-ray diffraction (XRD), reported in Ixer et al. (2019, 2020) and Bevins et al. (2020, 2022a, 2023a, 2023b). In view of these publications only a brief review of the Altar Stone is presented here.

The overall dimensions of the Altar Stone at Stonehenge and its geometry (measuring 4.9 m long by 1 m wide by 0.5 m thick) suggest that its original bed thickness must be > 50 cm, with widely spaced (~5 m) joint sets; the tabular nature suggests that the original bed geometry also has a tabular rather than strongly channelised or lenticular form. The detrital mineralogy of the Altar Stone is dominated by sub-angular to sub-rounded monocrystalline quartz grains showing straight extinction although rare, larger grains are strained. Plagioclase is much more abundant than K-feldspar, the latter comprising < 0.35 modal % of the rock (Bevins et al., 2020, 2022a, 2023b). Lithic grains (rock fragments) are the same size as the quartz and feldspar grains; most are internally fine-grained and include siliceous "cherts", polycrystalline metamorphic quartz, phyllite and fine-grained sandstone, along with rare, fine-grained graphic granite and quartz-chlorite intergrowths. Phyllosilicates are abundant (~5 %) with chlorite > muscovite > biotite. Heavy minerals, identified optically and through automated mineralogy and Raman spectroscopy, include Fe oxides, chromite, Ti oxides (rutile and anatase), rare ilmenite, apatite, garnet (mostly almandine and spessartine), tourmaline (mostly schorl) and variably metamict zircon. The diagenetic features of the Altar Stone are dominated by abundant (up to 18 %) pore filling and replacive calcite cement and late-stage pore filling baryte (~0.8 %).

2.2. Orkney stone circles

Stones comprising the Ring of Brodgar and the Stones of Stenness stone circles have been divided into a series of petrographic groups, which are described below (see Downes et al., 2019; Richards, 2019). Note, however, that the petrographic groups for the two stone circles are not the same (i.e. Stenness Group 1 is not the same lithology as Ring of Brodgar Group 1), and here we treat the two stone circles separately.

2.2.1. Stones of Stenness

The Stones of Stenness are commonly said to have been erected by the 30th century cal BC (Griffiths and Richards, 2019, 285) and form a stone circle *c.* 30 m in diameter and originally comprising 11 (or possibly 12) upright monoliths. Whilst some stones are represented by broken and buried stumps, others still stand 5 to 6 m in height. Four upright monoliths remain today, namely stones 2, 3, 5 and 7, whilst Stone 8 is exposed as a stump (see Fig. 2). Collins (1976) recorded the lithology of the Stones of Stenness, identifying five different groups (groups 1–5) representing nine stones (see Richards, 2019, Table 3.1).

- Group 1 (stones 2 and 3) are fine- to medium-grained, blue-grey sandstones showing ripple laminations and carious weathering.
- Group 2 (stones 5 and 7) are coarser-grained sandstones compared to stones 2 and 3 and are more siliceous and less micaceous. Being more siliceous these two stones are more resistant to weathering; and
- Group 3 (Stone 8) (Group 3) is a carbonate-rich siltstone with thin laminations and hence is prone to weathering.

It has been suggested that the Group 2 sandstones were sourced from Vestra Fiold (Richards et al., 2019, 128). In fact, similarities are also seen between the form and size of one of the monoliths excavated at Vestra Fiold to standing stones at both the Stones of Stenness and the Ring of Brodgar (see later).

2.2.2. Ring of Brodgar

The Ring of Brodgar is thought to be younger than the Stones of Stennes circle, dating to around 2600–2400 BCE (see Downes et al., 2019), broadly coincident with the second stage 2 of the construction of

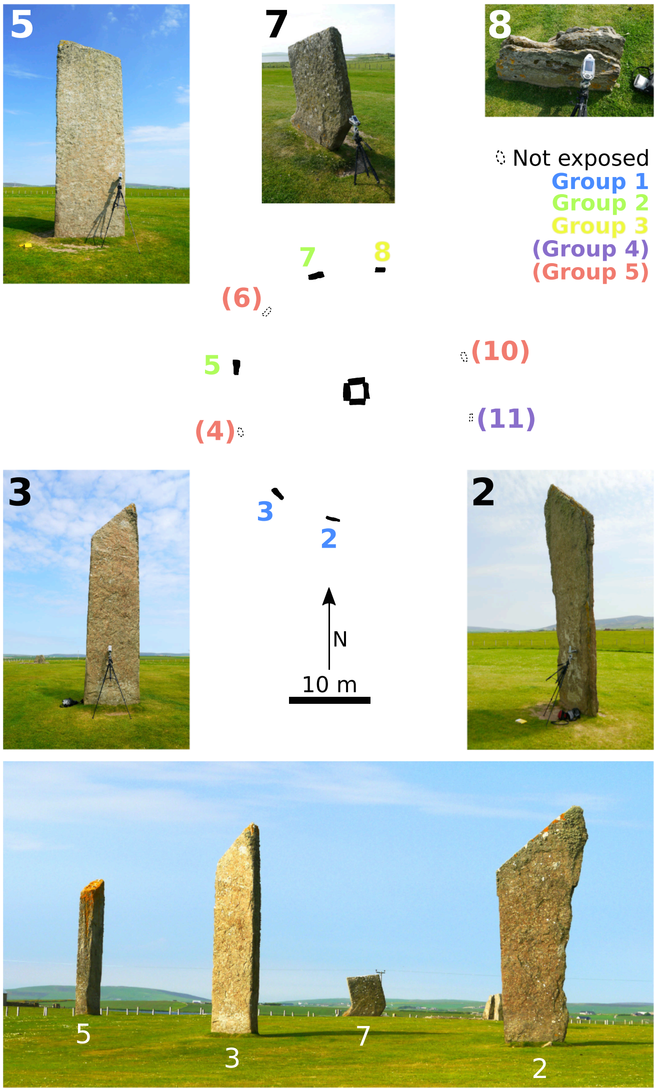

Fig. 2. Map of the Stones of Stenness, labelled with stone numbers colour-coded to indicate the petrographic groups presented in Richards (2019, Table 3.1. after Collins, 1976). Stones/Groups labelled with numbers in parentheses are not exposed. Photographs show each analysed stone, with an overview photograph of the site. All photographs by Nick Pearce.

Stonehenge (Parker-Pearson, 2023). The Ring of Brodgar has a diameter of *c.* 103 m and is the fourth largest stone circle in the British Isles (see Downes et al., 2019; Parker Pearson et al., 2021). Originally it might have comprised 60 monoliths but only 21 erect monoliths exist today, along with a number of broken stumps and fallen monoliths (see Fig. 3). Downes et al. (2019, Table 4.10) recorded seven stone groups, namely:

- Groups 1 and 2 are medium to thick bedded, fine-grained, grey-brown sandstones, thought to be derived from Vestra Fiold;
- Group 3 are thinly laminated, fine grained, grey-brown ‘flagstones’;
- Group 4 are laminated, fine-grained, grey-brown sandstones similar to groups 1 and 2 but with greater disturbance to its layering. It is thought to be sourced from Staneyhill;
- Group 5 are massive, laminated, grey-brown sandstones with iron oxide weathering;
- Group 6 is a laminated, golden-brown, iron-rich sandstone with mudstone layers; and
- Group 7 is a fine-grained, soft, originally yellow sandstone thought to belong to the Eday Group (the highest stratigraphic group in the Orkneys) and thought to be derived from Houton.

2.3. Old Red Sandstone Supergroup field samples

Thirteen samples were hand collected from strata belonging to the ORS Supergroup from across Mainland Orkney for comparison with the Stonehenge Altar Stone and also with the stones comprising the Stones of Stenness and the Ring of Brodgar stone circles. The geographical distribution of these field samples is illustrated in Fig. 1 and their stratigraphical and location details are presented in Table 1 and Fig. 4. The samples cover almost the full stratigraphical range of the ORS (Devonian) sequences exposed on Orkney.

2.4. Monoliths from the Neolithic quarry at Vestra Fiold

Although we did not analyse the recumbent monoliths at the Vestra Fiold quarry described and illustrated by Richards et al. (2019), below we compare the size of two monoliths (Monolith 1 and Monolith 2) from this site with the size of stones from the Ring of Brodgar and the Stones of Stenness circles and also with the size of the Altar Stone.

3. Stone size comparisons between the Altar Stone, standing stones of the Ring of Brodgar and the stones of Stenness, and monoliths from the Vestra Fiold quarry

Fig. 5 compares the size of the Altar Stone from Stonehenge to the standing stones of the Ring of Brodgar and the Stones of Stenness stone circles (using measurements in feet from the source documents, see Figure caption) and two monoliths from the Vestra Fiold quarry. Where stones are standing, they have been plotted as their above ground height, or for fallen stones, as their full length. To compare with the Altar Stone, the lengths of standing stones have been recalculated assuming that between a quarter and a fifth of their full length is currently buried to give a “calculated length”. The sizes of the two monoliths at Vestra Fiold are taken from Richards et al. (2019). The Altar Stone is a little longer and thicker (thickness is parallel to bedding) than the average Orcadian standing stone and Monolith 1 from Vestra Fiold sits within the range of their measurements (Monolith 2 has a very different size aspect to all other stones under investigation here). The aspect ratio of these stones will reflect the general bedding thickness and joint pattern in their source sandstones, and whilst in its own right this feature does not confirm a connection, it suggests a similar post-depositional tectonic regime to form the jointing in the sediments, and thus does not exclude the possibility of a relationship between these rocks.

4. Samples studied and methods

4.1. Petrographic and SEM-EDS investigations

Three samples were examined microscopically. Samples from both the Lower Stromness Flagstone Formation (5505 and 5516) and the Upper Stromness Flagstone Formation (sample 5510) were studied, these formations dominating the aerial extent of the ORS on Mainland Orkney. The Qui Ayre Sandstone Formation and the Eday Group were not investigated microscopically as XRD (see later) showed that these two units are highly siliceous and hence dissimilar to the Altar Stone. One of the samples from the Lower Stromness Flagstone Formation (sample 5505; also termed Yesnaby 1) was examined microscopically as it was shown by pXRF to have a high Ba content (see later) and by XRD to contain baryte and hence shows some initial similarity to the Altar Stone.

Optical microscopy was used to provide a qualitative assessment of the petrography and mineralogy of sampled rocks. Polished thin-sections 5505, 5510 and 5516 were investigated initially using a × 20 hand lens and Geo Supplies Ltd grain size card. The petrography of each section was analysed under plane- and cross-polarised transmitted light using a Carl Zeiss Amplival Pol u dual-purpose microscope (×6.3 and ×

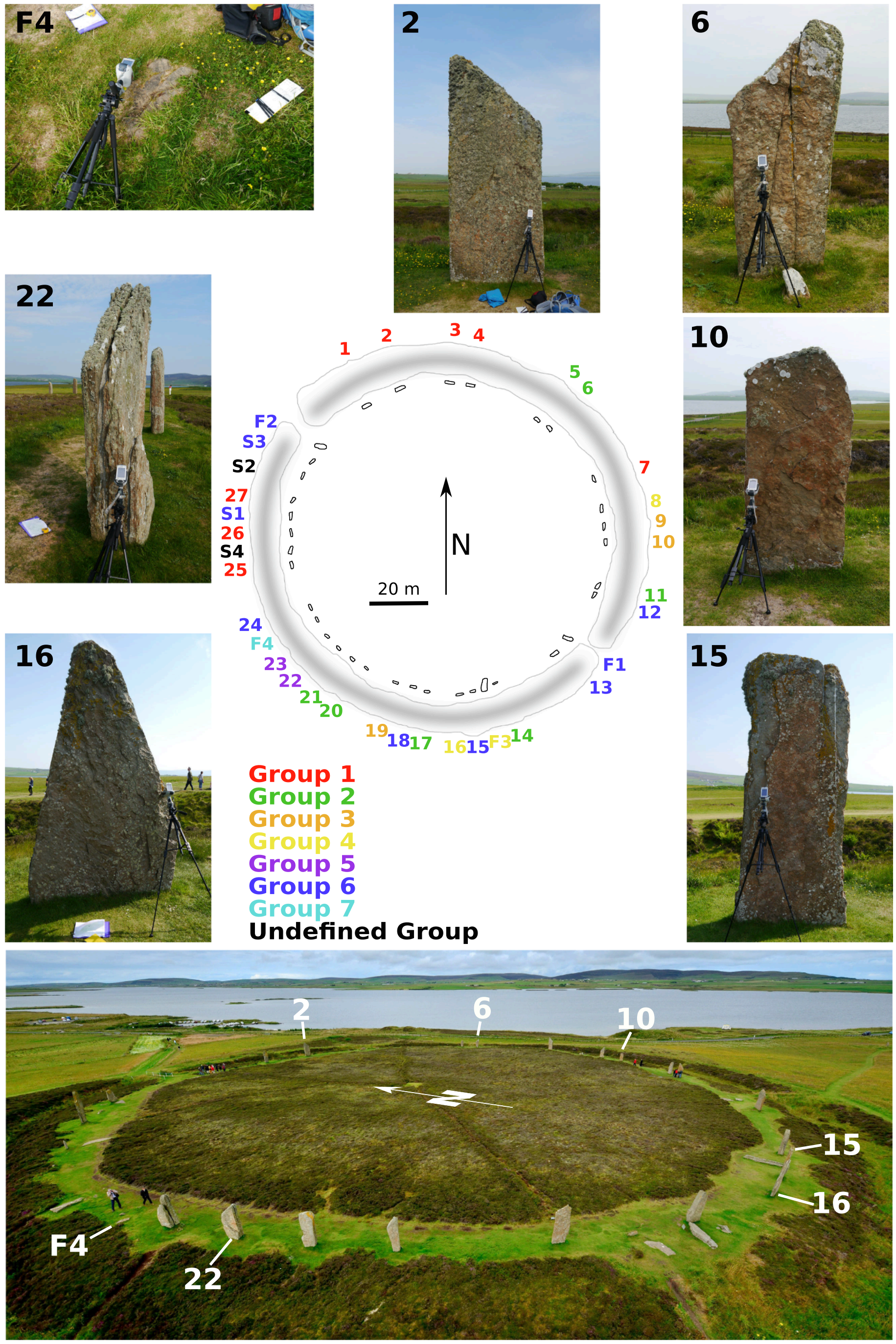

Fig. 3. Map of the Ring of Brodgar, labelled with stone numbers colour-coded to indicate the petrographic groups identified by Downes et al. (2019). Grey arcs indicate the ditch around the stone circle. Photographs show each analysed stone, taken from the inside of the Ring, looking outwards, except 22 looking SE along the circumference. The aerial photograph of the site shows the disposition of the analysed stones. Aerial photograph courtesy of Adam Sandford, reproduced with permission. All other photographs by Nick Pearce. Prefixes: F – fallen stone, S – stump.

12.5 objectives with × 12.5 eye pieces, giving overall magnifications of × 80 and × 155 respectively). Each section was then investigated in reflected light using a Zeiss Universal reflected light microscope (with × 4.5 air, ×16 oil and × 40 oil immersion lens). Mineral identification in transmitted and reflected light was made following standard optical mineralogy texts and atlases, with petrographic descriptions also following standard protocols for sedimentary rocks. All mineral phases greater than 2 μm diameter were identified. Note, however, that the fine-grained nature of the TiO_2 phases present sometimes prevented further discrimination. Where a TiO_2 phase could be identified with certainty it is given a mineral name, where not, it is simply referred to as a 'TiO_2 mineral'.

Following petrographic analysis and imaging, the three thin sections were carbon coated and then analysed and imaged using automated scanning electron microscopy with linked energy dispersive spectrometers (SEM-EDS). Analysis was undertaken using a Hitachi SU3900 scanning electron microscope fitted with a single large area (60 mm^2) Bruker SDD energy dispersive spectrometer and running the AMICS automated mineralogy package. Beam conditions were optimised for analysis and therefore an accelerating voltage of 20 kV coupled with a beam current of approximately 15 nA was used. All samples were measured using the same analytical parameters and with a segmented

Table 1
A list of Old Red Sandstone field samples used in this study along with sampling sites and stratigraphic data.

Locality	Sample number	Latitude/ Longitude	Nat Grid Ref	Stratigraphic horizon
Yesnaby 1	5505	59° 01′ 28.603″N 3° 21′ 35.729″W	HY 22026 16079	Lower Stromness Flagstone Formation
Yesnaby 2	5506	59° 00′ 28.033″N 3° 21′ 39.583″W	HY 21926 14207	Lower Stromness Flagstone Formation
Yesnaby 3	5507	59° 01′ 05.750″N 3° 21′ 38.526″W	HY 21967 15373	Qui Ayre Sandstone Formation
Marwick 1	5508	59° 05′ 47.294″N 3° 20′ 52.200″W	HY 22881 24066	Upper Stromness Flagstone Formation
Birsay 1	5509	59° 08′ 08.584″N 3° 19′ 31.331″W	HY 24255 28410	Upper Stromness Flagstone Formation
Birsay 2	5510	59° 08′ 08.584″N 3° 19′ 31.331″W	HY 24255 28410	Upper Stromness Flagstone Formation
Gurness 1	5511	59° 07′ 26.141″N 3° 05′ 03.624″W	HY 38026 26848	Upper Stromness Flagstone Formation
St Nicholas Kirk 1	5512	58° 53′ 32.507″N 2° 51′ 29.110″W	HY 50646 00875	Caithness Flagstone Group
St Nicholas Kirk 2	5513	58° 53′ 32.507″N 2° 51′ 29.110″W	HY 50646 00875	Caithness Flagstone Group
Bu 1	5514	58° 55′ 01.148″N 3° 08′ 27.274″W	HY 34394 03860	Eday Group
Netherton Road 1	5515	58° 57′ 07.686″N 3° 18′ 54.637″W	HY 24436 07958	Lower Stromness Flagstone Formation
Netherton Road 2	5516	58° 57′ 07.686″N 3° 18′ 54.637″W	HY 24436 07958	Lower Stromness Flagstone Formation
Skipy Geo 1	5517	59° 08′ 16.665″N 3° 18′ 46.768″W	HY 24968 28646	Upper Stromness Flagstone Formation

field image mode of analysis. This analytical mode subdivides the BSE image into domains (segments) of similar brightness which represent different mineral grains/crystals and then acquires a representative EDS X-ray spectrum from a point within the segment; the mineral identified is then assigned to the entire segment. Measurements are optimised to highlight both textural and modal mineralogical information and so an effective image resolution of 2.48 μm is achieved. The results from the automated SEM-EDS analysis are directly comparable with previous datasets from samples of the Altar Stone.

4.2. X-ray diffraction

All thirteen field samples from the ORS collected on Mainland Orkney were analysed by X-ray diffraction. Sample location and stratigraphic details are provided in Fig. 1 and Table 1. The samples cover the main stratigraphic units on Mainland Orkney. Three samples (MS1, MS2 and MS3) of debitage from the Altar Stone were also analysed.

Portions of the analysed samples were gently disaggregated and crushed to a powder using an agate mortar and pestle. A subsample of ≈3 g was then transferred to a McCone mill, milled for 12 min in ethanol using zirconia milling elements, and the resulting slurry spray dried directly from the mill to produce a random powder specimen as described by Hillier (1999). The powders were loaded into circular aluminium holders and run on a Bruker D8 advance X-ray diffractometer from 4 to 70° 2Θ, counting for 96 s per 0.0196° step using a Lynxeye XE position sensitive detector and Ni-filtered Cu radiation. Quantitative mineralogical analysis of the random powder patterns was made using the powdR package (Butler and Hillier, 2021a) using procedures similar to those described in Butler and Hillier (2021b). In essence, the method of quantitative analysis is a full pattern fitting method, wherein minerals are first identified by routine search match procedures and selected for inclusion in the pattern fitting, the approach being repeated until all minerals are identified and the fitted pattern, which is the weighted sum of library patterns, is a close match to the measured pattern.

Additionally, a further portion of each sample was suspended and dispersed in deionised water and a < 2 μm clay size fraction separated by timed sedimentation according to Stokes' law. Clay fractions were then mounted on glass slides by a filter peel method and run on the D8 advance XRD from 2.5 to 45° 2Θ, counting for 16.5 s per 0.0196° step. The clay specimens were run three times, first following drying in air, secondly after solvation with ethylene glycol by vapour pressure overnight, and thirdly following heating to 300 °C for one hour on a hotplate. Clay minerals were identified and by comparison the responses to the various treatments and semi-quantitative estimates of clay mineral composition (relative wt. %) were made following procedures outlined in Hillier (2003).

4.3. Heavy mineral analysis

Five samples from the ORS sequences on Mainland Orkney, covering the main successions, were selected for heavy mineral analysis. These were samples 5507, 5508, 5512, 5514 and 5516. Table 1 and Fig. 1 provide location and stratigraphic details for these samples.

Initially, each sample was mechanically disaggregated using a manual press to create small rock chips of rock. This was followed by manual disaggregation using an agate mortar and pestle, conducted wet so as to reduce grinding of the constituent mineral grains. The products of the disaggregation were then first wet sieved, using a 500 μm sieve, and then using a 15 μm nylon mesh to obtain material with a grain size ranging from 15-500 μm. To obtain a heavy mineral separate, this fraction was then centrifuged in a solution of SPT (sodium-polytungstate) with a density of 2.90 g/cm^3. In order to recover the heavy and light fraction from each sample, a partial freezing method with liquid nitrogen was used. Grain mounts of the heavy minerals were prepared using an aliquot of the separated heavy minerals, prepared using a micro-splitter, and then mounted in Canada Balsam (refractive index n = 1.56) on a glass side on a hot plate. This preparation method follows the protocol described by Andò (2020).

Identification of minerals was undertaken in grain mounts by the use of an optical microscope and by Raman spectroscopy. A Renishaw inVia spectrometer, equipped with a 532 nm green laser and a 50x long working distance objective, enabled recognition of mineral grains, including varieties. Raman spectra were compared with an in-house data base and also with the Raman spectra identified in sample MS1 (reported in Bevins et al., 2023a).

4.4. Portable XRF

In previous studies we have reported on portable XRF (pXRF) analyses of the Altar Stone (*in situ* analyses performed on two separate visits), analyses of six small pieces of debitage (which were confirmed to be fragments of the Altar Stone; Bevins et al., 2022a), and of sample 2010 K 240 from the collections of Salisbury Museum (sometimes referred to as Wilts 277), which we confirmed the authenticity of as a piece collected from the underside of the Altar Stone in 1844 (Bevins et al., 2023b).

In this current study, analyses were performed to cover each of the lithological groups described above for stones from the Ring of Brodgar and the Stones of Stenness stone circles. At the Stones of Stenness, all the exposed stones were analysed, these belonging to the three petrographic groups *viz.* stones 2, 3 (both Group 1), 5, 7 (both Group 2) and the stump of 8 (Group 3) (see Fig. 2). Stones from groups 4 and 5 at Stenness are currently not exposed. At the Ring of Brodgar, seven stones were

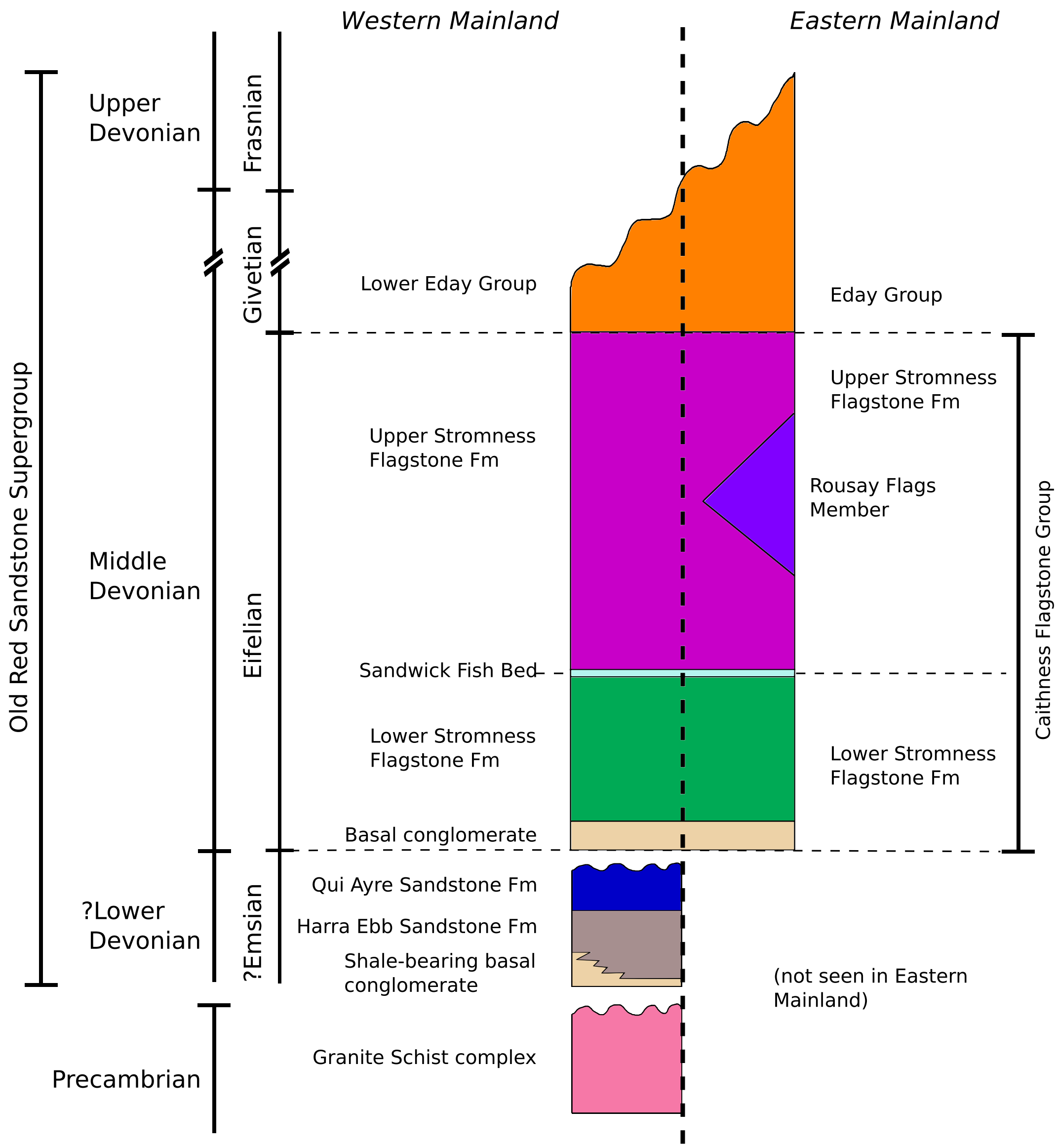

Fig. 4. Stratigraphy of the area around the Ring of Brodgar and Stones of Stenness on Mainland, Orkney. Colours used are the same as those used in Figs. 9 and 10. Simplified from British Geological Survey (1999), Orkney Islands, Scotland Special Sheet (solid and drift), 1:100 000.

analysed *viz.* stones 2 (Group 1), 6 (Group 2), 10 (Group 3), 16 (Group 4), 22 (Group 5), 15 (Group 6), and the poorly exposed fallen stone F4 (Group 7) (see Fig. 3), these representing one from each of the seven groups described in Downes et al. (2019). Note that the defined petrographic groups for the two stone circles ***are not*** the same.

Portable X-ray fluorescence analyses were conducted on the standing stones at the Stones of Stenness and Ring of Brodgar sites, and at a series of sites exposing the geology of predominantly western Mainland, Orkney, during uncommonly warm and dry weather between 12 and 15th June 2023. Analyses were performed to compare with the *in situ* Stonehenge Altar Stone and debitage derived therefrom (Bevins et al., 2022a; Bevins et al., 2023b).

In addition, all thirteen field samples from the ORS samples collected were analysed by pXRF. Sample location and stratigraphic details are provided in Fig. 1 and Table 1. These cover the main stratigraphic units on Mainland Orkney.

All analyses were performed using a Thermo Fisher Scientific™ Niton™ XL3t Goldd + handheld XRF analyser. The Niton pXRF uses a 2 W Ag anode X-ray tube, which can operate at between 6–50 kV and 0–200 μA, operating conditions being varied during the "TestAllGeo" analysis method. The instrument determines a range of elements in geological materials from Mg to U by using, in sequence, different filters to optimise sensitivity. Light element analyses (e.g. Mg, Al, Si, S) are less accurate without flushing the instrument with He, and these are also particularly sensitive to the presence of moisture within or on the surface of the sample. For all analyses, the total acquisition time was 100 s, divided between four operating modes (Main range 30 s, Low range 30 s, High Range 20 s, Light range 20 s) using an 8 mm diameter analysis spot, giving an analysed surface area of ~ 50 mm^2. Spectra were collected on a silicon drift detector, which are processed and calibrated by the manufacturers installed calibration. Here, across four days, we performed around 20 analyses on each standing stone (with the exception the fallen stone F4 at Brodgar and the stump of Stenness Stone 8), and between five to ten analyses of the weathered surfaces of each ORS field site. Instrument calibration was monitored using a piece of the Big Obsidian Flow from the Newbery Volcano in Oregon. All analyses are presented in the Supplementary Material 1. Elsewhere we have discussed at length analytical methods and instrument accuracy and precision, and these aspects of the method are not revisited here (but see Bevins et al., 2022a; Bevins et al., 2022b; Pearce et al., 2022; Bevins

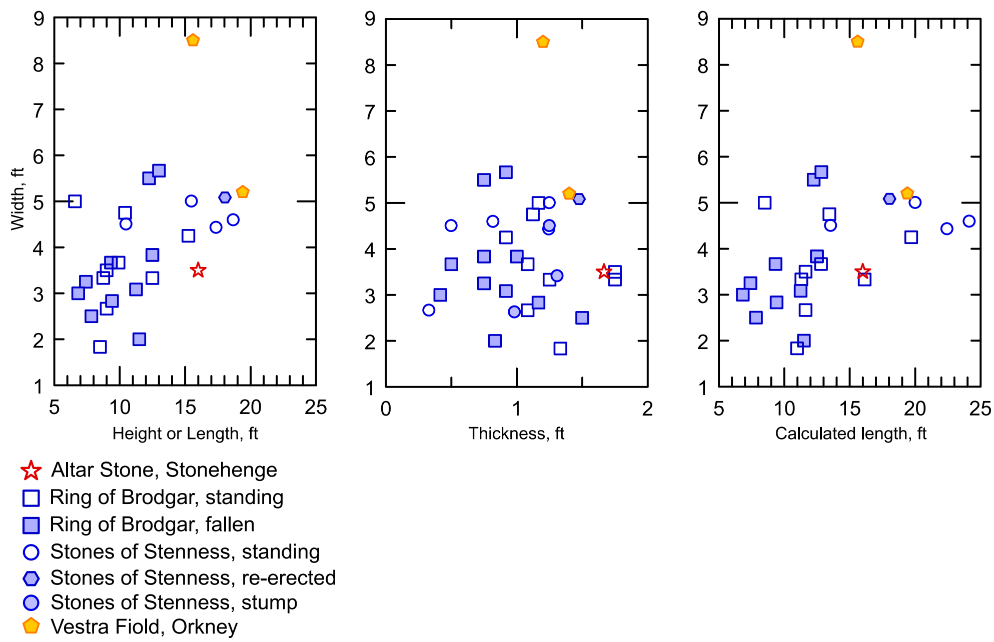

Fig. 5. Dimensions of the Altar Stone, Stonehenge, compared with stones from the Ring of Brodgar and the Stones of Stenness (including single standing stones in the vicinity) on Orkney. Dimensions in feet (ft, 1 foot = 0.3048 m) taken from documents of the Records of the Society of Antiquaries of Scotland, Edinburgh, Scotland, available online at http://canmore.org.uk/collection/ from collection numbers for the Ring of Brodgar – 1322798, 1327303; the Watchstone, Stenness – 1322793, 1327303; and the Stones of Stenness from Ritchie et al. (1978) and https://canmore.org.uk/collection/1322798. "Height" in the upper panel is the measured above ground height for standing stones, this has been converted to an approximate "Calculated length" by adding 7/24ths to the above ground height of standing stones assuming that between 1/4 to 1/5 of the stone is buried. For fallen stones, "calculated length" is the full, measured length of the stone. Stumps are excluded from the height/length graphs.

et al., 2023a; Bevins et al., 2023b).

5. Results

5.1. Petrography and automated SEM-EDS

Detailed petrographic accounts of the three samples examined microscopically are presented in Supplementary Material 2. Brief petrographic summaries are presented here along with the quantitative modal mineralogy based on automated SEM-EDS analysis.

In thin section, Sample 5516 (Lower Stromness Flagstone Formation) is a very well sorted, fine-grained, laminated, dolomitic, micaceous, arkosic sandstone. Dominant quartz is accompanied by muscovite laths and minor amounts of plagioclase, untwinned feldspar and microcline, all set within a rhombic carbonate cement. Thin heavy mineral laminae comprise Fe-Ti oxides (now pseudomorphed by titania), zircon, rutile,

Table 2
Modal mineralogy (area %) for samples 5505 and 5516 from the Lower Stromness Flagstone Formation and Sample 5510 from the Upper Stromness Flagstone Formation measured using automated SEM-EDS.

Sample Number	5505	5516	5510
Formation	Lower Stromness Flagstone	Lower Stromness Flagstone	Upper Stromness Flagstone
Quartz	53.50	57.42	44.53
K-feldspar	13.98	15.38	6.72
Plagioclase	15.14	9.53	18.44
Muscovite	2.52	2.37	1.68
Biotite	0.38	0.08	0.68
Kaolinite	0.02	0.01	0.01
Chlorite	0.62	0.17	0.49
Illite	3.52	2.65	4.46
Ferroan Illitic Clays	0.21	0.01	0.50
Calcite	0.13	0.33	18.05
Dolomite	1.92	0.75	1.12
Ferroan Dolomite	5.76	10.28	0.11
Siderite	0.53	0.29	0.10
Fe Oxides	0.20	0.17	2.42
Chromite	0.00	0.00	0.01
Baryte	1.07	0.00	0.00
Ilmenite	0.00	0.00	0.01
Ti Oxides	0.15	0.22	0.29
Pyrite	0.01	0.01	0.00
Apatite	0.05	0.17	0.25
Garnet	0.01	0.00	0.01
Tourmaline	0.01	0.01	0.02
Zircon	0.06	0.04	0.05
Undifferentiated	0.19	0.08	0.05

authigenic tabular titania, apatite, tourmaline and chromite. Trace amounts of sulphides, including marcasite and lesser amounts of pyrite, chalcopyrite and sphalerite are also present. The sandstone is tightly compacted, with sutured grain contacts and kinked micas. The modal mineralogy is shown in Table 2; in this summary major phases are > 10 %, minor 1–10 % and trace < 1 % by area. Sample 5516 is dominated by major detrital quartz and K-feldspar along with ferroan dolomite cement (Fig. 6). Minor minerals present are plagioclase, muscovite and illite along with trace biotite, kaolinite, chlorite, ferroan illite, calcite, dolomite, siderite, Fe oxides, Ti oxides, pyrite, apatite, tourmaline, zircon and "undifferentiated" minerals (Table 2).

Sample 5505 (Lower Stromness Flagstone Formation) is a well-sorted, dolomitic, micaceous, arkosic, fine-grained sandstone with carbonate-muscovite rich laminae. Quartz is accompanied by muscovite laths and plagioclase, untwinned feldspar and microcline; much feldspar shows euhedral terminations, and all are set within a rhombic dolomitic cement. Rare carbonate bioclasts are also present. Poorly developed heavy mineral laminae comprise iron titanium oxides (now pseudomorphed by titania) and zircon, with lesser amounts of rutile, apatite, tourmaline and chromite (Fig. 6). There are trace amounts of pyrite and very locally baryte forms a cement about quartz grains and is also associated with small-scale fractures (Fig. 6). Sample 5505 is dominated by major detrital quartz, plagioclase and K-feldspar. Minor minerals present are muscovite, illite, dolomite, ferroan dolomite and baryte, along with trace biotite, kaolinite, chlorite, ferroan illite, calcite, siderite, Fe oxides, Ti oxides, pyrite, apatite, garnet, tourmaline, zircon and "undifferentiated" minerals (Table 2).

Sample 5510 (Upper Stromness Flagstone Formation) is a fine-grained, ripple cross laminated, calcitic, micaceous, arkosic sandstone with carbonate-muscovite rich laminae. Laminae are defined by grain size variation (Fig. 6). Quartz is accompanied by muscovite laths and plagioclase, untwinned feldspar and microcline; much feldspar shows euhedral terminations, and all are set within a calcite cement. Poorly developed heavy mineral laminae comprise iron titanium oxides now pseudomorphed by titania, and zircon, with minor titania, apatite, tourmaline and chromite. Trace amounts of pyrite and authigenic tabular rutile clusters are present. At thin section scale oval trace fossils are observed. Sample 5510 is dominantly composed of major detrital quartz and plagioclase with a calcite cement. Minor minerals present are K-feldspar, muscovite, illite, dolomite and Fe oxides, along with trace biotite, kaolinite, chlorite, ferroan illite, ferroan dolomite, siderite, chromite, ilmenite, Ti oxides, apatite, garnet, tourmaline, zircon and "undifferentiated" minerals (Table 2).

5.2. X-ray diffraction

Bulk mineralogical compositions (wt. %) of the samples are given in Table 3 while Table 4 gives the clay mineral compositions. Minerals identified and quantified in the bulk samples include, quartz, plagioclase feldspar, K-feldspar, calcite, dolomite/ankerite, pyrite, baryte, halite,

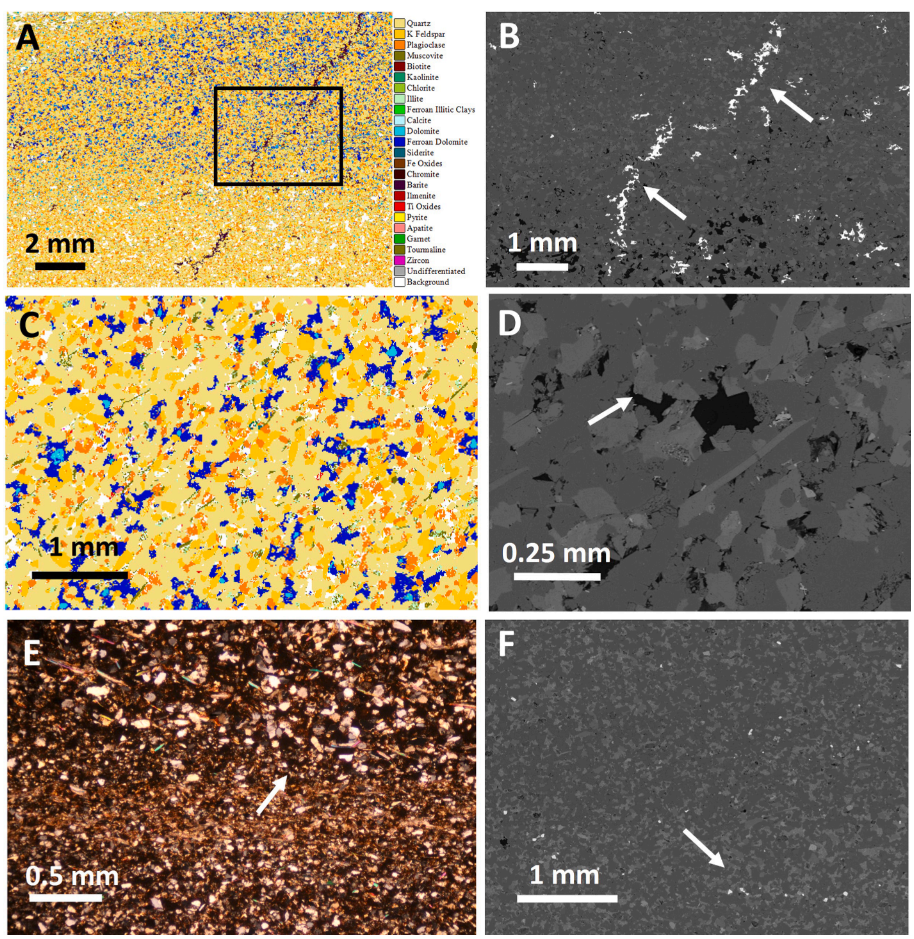

Fig. 6. Petrography of the examined Orkney sandstones. (A) Automated SEM-EDS false colour image of Sample 5505. Note the patchy distribution of the dolomite cements and the cross-cutting baryte filled fracture. (B) SEM backscatter electron (SEM-BSE) image of Sample 5505; the bright phase (arrowed) is baryte; area of image is that shown in the inset box in (A). (C) Automated SEM-EDS false colour image of Sample 5516, with pore filling dolomite cements. (D) SEM-BSE image of Sample 5516; note the abundant pore filling rhombic dolomite cements (arrowed). (E) Cross polarised light image of Sample 5510; note lamination defined by grain size variation (arrowed). (F) SEM-BSE image of Sample 5510; laminae defined by heavy minerals arrowed. Samples 5505 and 5516 from the Lower Stromness Flagstone Formation. Sample 5510 from the Upper Stromness Flagstone Formation.

Table 3
Bulk mineralogical composition (wt. %) determined by XRD for Old Red Sandstone field samples used in this study.

Stratigraphy	Hutton ID	Sample no.	Quartz	Plagioclase	K-feldspar	Calcite	Dolomite/ Ankerite	Pyrite	Baryte	Halite	Goethite	Muscovite (2 M1)	I+I/S-ML	Chlorite (Tri)	Chlorite (Di)	Kaolinite	Total
Lower Stromness Flags Fm	1,388,209	5505	54.5	14.5	11.1	0.1	8.3	nd	2	nd	3.7	2.1	3.7	nd	nd	nd	100
Lower Stromness Flags Fm	1,388,210	5506	41.5	13.7	17.9	2.9	2.8	nd	nd	0.1	8.2	5.1	7.8	nd	nd	nd	100
Qui Ayre Sandstone Fm	1,388,211	5507	83.6	0.1	5.9	nd	nd	nd	0.2	0.2	1.6	1.1	7.3	nd	nd	nd	100
Upper Stromness Flags Fm	1,388,212	5508	26.6	18.7	8.7	8.7	2.4	1.1	nd	0.2	2.5	6.8	11.5	12.8	nd	nd	100
Upper Stromness Flags Fm	1,388,213	5509	12.2	15.9	5.2	16.5	8.4	0.7	nd	0	1.7	8.1	16.3	15	nd	nd	100
Upper Stromness Flags Fm	1,388,214	5510	38.9	19.1	6.3	15.2	0.8	nd	nd	nd	6.9	4	8.8	nd	nd	nd	100
Upper Stromness Flags Fm	1,388,215	5511	13.4	11.1	3.1	40.4	13.5	1.1	nd	0.1	1.8	5.1	10.4	nd	nd	nd	100
Caithness Flagstone Group	1,388,216	5512	43.3	28.2	10.2	0.2	nd	nd	nd	0.3	4.4	3.5	7.6	2.3	nd	nd	100
Caithness Flagstone Group	1,388,217	5513	37.6	31.1	11.1	6.1	1.3	nd	nd	0.1	2.4	2.2	5.4	2.7	nd	nd	100
Eday Group	1,388,218	5514	82.3	0.2	5.2	nd	nd	nd	nd	nd	0.8	0.8	3.8	nd	nd	6.9	100
Lower Stromness Flags Fm	1,388,219	5515	45.3	14.4	16.7	nd	0.3	nd	nd	nd	3.5	8.6	11.2	nd	nd	nd	100
Lower Stromness Flags Fm	1,388,220	5516	57.2	11	12.8	1.9	5.6	nd	nd	0	4.5	3	4	nd	nd	nd	100
Upper Stromness Flags Fm	1,388,221	5517	37.6	28.2	7.5	2.8	7.4	nd	nd	0.1	1.5	3.8	6.2	4.9	nd	nd	100
Altar Stone	1,307,438	MS1	55.1	10.6	nd	13.2	nd	nd	1.1	nd	nd	1.1	11.4	3.3	0.7	3.5	100
Altar Stone	1,307,439	MS2	54.9	10.7	nd	13.1	nd	nd	1.1	nd	nd	1.1	11.5	3.3	0.7	3.6	100
Altar Stone	1,307,440	MS3	54.4	10.7	nd	14.2	nd	nd	0.8	nd	nd	1.1	11.1	3.3	0.6	3.8	100

Table 4
Relative percentage of clay minerals in the < 2 μm clay size fraction© determined by XRD for Old Red Sandstone field samples used in this study.

Hutton ID	Sample no.	Sample ID	Illite©	Chlorite(Tri) ©	Corrensite (Tri) ©	Chlorite (Di) ©	Tosudite ©	I/S-ML©	Smectite(Di) ©	Kaolinite©
1,388,209	5505	**Lower Stromness Flags Fm**	100	–	–			–	–	–
1,388,210	5506	**Lower Stromness Flags Fm**	100	–	–			–	–	–
1,388,211	5507	**Qui Ayre Sandstone Fm**	100	–	–			–	–	–
1,388,212	5508	**Upper Stromness Flags Fm**	44	19	37			–	–	–
1,388,213	5509	**Upper Stromness Flags Fm**	64	36	–			–	–	–
1,388,214	5510	**Upper Stromness Flags Fm**	100	–	–			–	–	–
1,388,215	5511	**Upper Stromness Flags Fm**	100	–	–			–	–	–
1,388,216	5512	**Caithness Flagstone Group**	74	7	–			–	20	–
1,388,217	5513	**Caithness Flagstone Group**	49	6	45			–	–	–
1,388,218	5514	**Eday Group**	10	–	–			49	–	41
1,388,219	5515	**Lower Stromness Flags Fm**	100	–	–			–	–	–
1,388,220	5516	**Lower Stromness Flags Fm**	100	–	–			–	–	–
1,388,221	5517	**Upper Stromness Flags Fm**	72	28	–			–	–	–
1,307,438	MS1	**Altar Stone**	14	5	–	13	21	26	–	21
1,307,439	MS2	**Altar Stone**	15	5	–	12	16	27	–	25
1,307,440	MS3	**Altar Stone**	19	5	–	12	15	33	–	16

goethite, muscovite mica, dioctahedral illite plus mixed-layer illite/smectite, trioctahedral chlorite plus/minus chlorite/smectite and kaolinite. Baryte was only positively identified in one sample from the Lower Stromness Flags at Yesnaby (sample 5505), though a trace was indicated in the underlying Qui Arye Sandstone also from Yesnaby (sample 5507). Clay minerals identified in the < 2 μm clay fractions include illite, (trioctahedral) chlorite, (trioctahedral) chlorite/smectite, R1 ordered illite/smectite, kaolinite and dioctahedral smectite. Key characteristics of the mineralogy of the three Altar Stone samples analysed are the presence of baryte and only trace amounts of K-feldspar, as reported previously by Bevins et al. (2022a, 2023a). In addition, the XRD data reported here show the presence of the regularly interstratified dioctahedral chlorite/dioctahedral smectite mineral tosudite which is present in the Altar Stone samples, but not the samples analysed from

Table 5
Qualitative summary description of the heavy mineral assemblages from Mainland Orkney Old Red Sandstone samples, compared with MS-1, a fragment of debitage from the Altar Stone, Stonehenge (data from Bevins et al., 2023a). Bold text: Most abundant heavy mineral, Underlined text: second most abundant mineral.

Sample: number, location	**5507 Yesnaby 3**	**5508 Marwick 1**	**5512 St Nicholas Kirk 1**	**5514 Bu 1**	**5516 Netherton Road 2**	**MS1 Altar Stone debitage**
Stratigraphy (from BGS GeoIndex)	Qui Ayre Sandstone Formation	Upper Stromness Flagstone Formation	Caithness Flagstone Group	Eday Group Sandstone	Lower Stromness Flagstone Formation	
15–500 μm (g)	22.6781	15.9786	13.8926	22.6953	11.2898	0.1453 (2–500 μm)
Light minerals (g)	22.5919	15.6600	13.7549	22.5943	10.9282	0.1334
Heavy minerals (g)	0.0262	0.1778	0.0587	0.0300	0.2848	0.0020
%weight heavy minerals	**0.1**	**1.1**	**0.4**	**0.1**	**2.5**	**1.4**
Transparent heavy minerals	Zircon; Rutile; Tourmaline (schorl and dravite); Cr-spinel (red); Clinozoisite (rare)	Apatite; Zircon; Tourmaline (schorl); Rutile; Cr-spinel (red)	Apatite; Zircon; Tourmaline (schorl); Rutile; Cr-spinel (red)	Zircon; Tourmaline (schorl); Rutile	Apatite; Zircon; Rutile (rare); Tourmaline (rare, dravite and schorl)	Apatite; Zircon, Rutile; Garnet; Tourmaline (schorl and dravite); Cr-spinel; Hornblende; Epidote
Opaque heavy minerals	Yes (unidentified)	**Pyrite (dominant)**	Not recognised	Yes (rare, unidentified)	Not recognised	Yes (rare, unidentified)
Semi-opaque heavy minerals	Fe-hydroxides; Ti-oxides	Fe-hydroxides; Ti-oxides	Ti-oxides; Fe-hydroxides (rare)	**Ti-oxides;** Fe-hydroxides	Ti-oxides (rare)	Ti-oxides
Carbonates	Not recognised	Yes	Not recognised o	Not recognised	**Yes (dominant)**	Yes
Authigenic heavy minerals	**Baryte (dominant);** Anatase	Anatase	Anatase	Anatase	Anatase	**Baryte;** Anatase
Phyllosilicates	Chlorite (rare)	Biotite and Chlorite (both with inclusions and subrounded)	**Biotite** and Chlorite (both subrounded)	Yes (rare)	Not recognised	Chlorite Biotite

Orkney.

5.3. Heavy minerals analysis

Table 5 provides a summary of the heavy mineral (HM) assemblages observed in the ORS samples from Mainland Orkney, along with the HM suite described in Altar Stone debitage sample MS1 (see Bevins et al., 2022a, 2023b). Minerals were determined optically and also by Raman spectroscopy. The main features are described here and illustrated in Fig. 7.

The HM suite in the Qui Ayre Sandstone Formation sample (5507) is dominated by baryte and zircon, associated with lesser amounts of Ti-oxides (anatase and rutile), tourmaline (schorl and dravite), red Cr-spinel and rare clinozoisite. In contrast, the Lower Stromness Flagstone Formation sample 5516 is dominated by carbonates, with rarer apatite, zircon, tourmaline (dravite and schorl) and Ti-oxides (rutile and anatase). The HM suite in Upper Stromness Flagstone Formation sample 5508 is characterized by abundant pyrite and is rich in sub-rounded flakes of biotite and chlorite (both with opaque inclusions), as well as apatite, zircon, tourmaline (schorl), red Cr-spinel, Fe-hydroxides and Ti-oxides (rutile and anatase). The undifferentiated Caithness Flagstone Group sample 5512 carries abundant biotite, along with apatite, zircon, tourmaline (schorl), red Cr-spinel, chlorite, Ti-oxides (rutile and anatase) and rare Fe-hydroxides. Finally, the Eday Group sample 5514 has a poorly sorted HM suite dominated by authigenic and detrital Ti-oxides (anatase and rutile), along with rarer zircon, tourmaline (schorl) and Fe-hydroxides. As has been reported previously (see above), the Altar Stone sample MS1 has abundant baryte, along with apatite, zircon, tourmaline (schorl and dravite), Cr-spinel, epidote, hornblende, Ti-oxides, chlorite and biotite. There are clear differences between the HM suite in the Altar Stone sample and those from the Orkney ORS samples.

5.4. Portable XRF

Initially, we review whether the composition of the Altar Stone compares to the compositions of stones from the Stones of Stenness and Ring of Brodgar stone circles. Fig. 8 shows a range of bivariate plots for the compositional data from the Stonehenge Altar Stone and the Stones of Stenness and Ring of Brodgar, for Ba, Sr, Rb and K, elements which were used (with the exception of K) by Bevins et al. (2023b) to show the compositional differences between possible ORS sources in the Anglo-Welsh Basin and the Altar Stone. Whilst the Stones of Stenness and those analysed from the Ring of Brodgar occupy broadly the same compositional space on these plots, it is clear for these elements that there is no compositional similarity between the Orcadian standing stones and the Stonehenge Altar Stone. These data, along with the mineralogical analyses, rule out any possibility of a correlation. Barium is considerably higher in the Altar Stone, which contains baryte (Bevins et al., 2020; Bevins et al., 2022a; Bevins et al., 2023a; Bevins et al., 2023b), whilst K and Rb are higher in the Orcadian samples, and although Sr concentrations from the Orkney stones are similar to the Altar Stone in their lower range (around 400 ppm), these extend to considerably higher concentrations (up to 1260 ppm). A solitary analysis from an Orkney stone (from 206 analyses), Stenness Stone 2, showed Ba in excess of the Altar Stone analyses (analysis 16, 13000 ppm Ba, see Fig. 8 and supplementary information), which probably relates to analysis of either a detrital grain of baryte in the sediment or a thin baryte-mineralised vein in that orthostat. Of the remaining Orkney standing stone analyses, only 5 exceed 1000 ppm Ba (from Brodgar groups 6 and 7, with up to 1260 ppm) and barely encroach on the range of Ba shown by the Altar Stone. These stones also have higher Sr/Ba ratios as well as notably higher Rb and K than the Altar Stone, and again cannot be correlatives.

Fig. 9 shows the compositional data for the Altar Stone and the stones from Stenness and Brodgar, plotted as triangular diagrams for Ba-Rb-Sr.

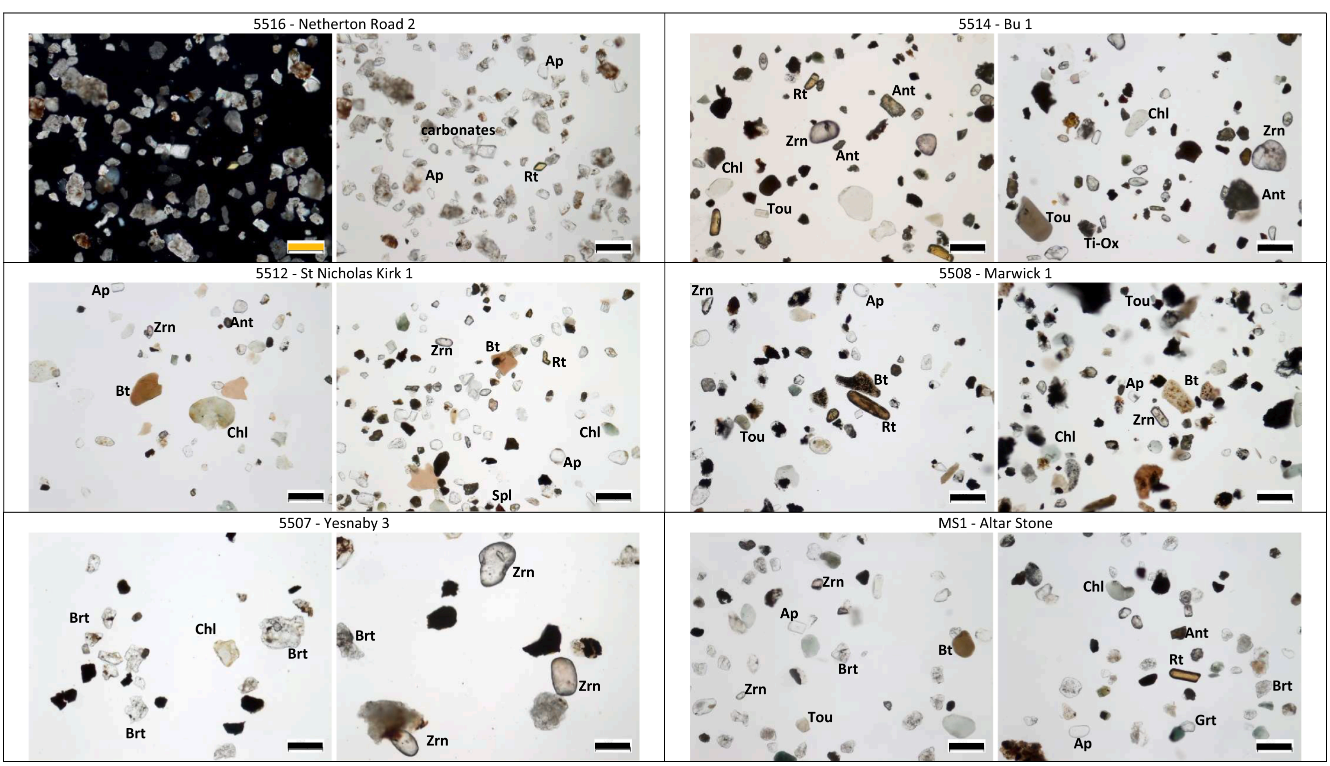

Fig. 7. Heavy mineral assemblages from five different Orkney formations are compared with the Altar Stone debitage samples MS-1. The **Fig.** shows two photos in transmitted light for each sample. Magnification is 10x and scale bar is 100 μm in all photos, with all images in plane polarized light excluding the left photo of sample 5516 (cross-polarized light). Abbreviations: Ant = anatase, Ap = apatite, Brt = baryte, Bt = biotite, Chl = chlorite, Grt = garnet, Rt = rutile, Spl = spinel, Ti-Ox = titanium oxides, Tou = tourmaline, Zrn = zircon.

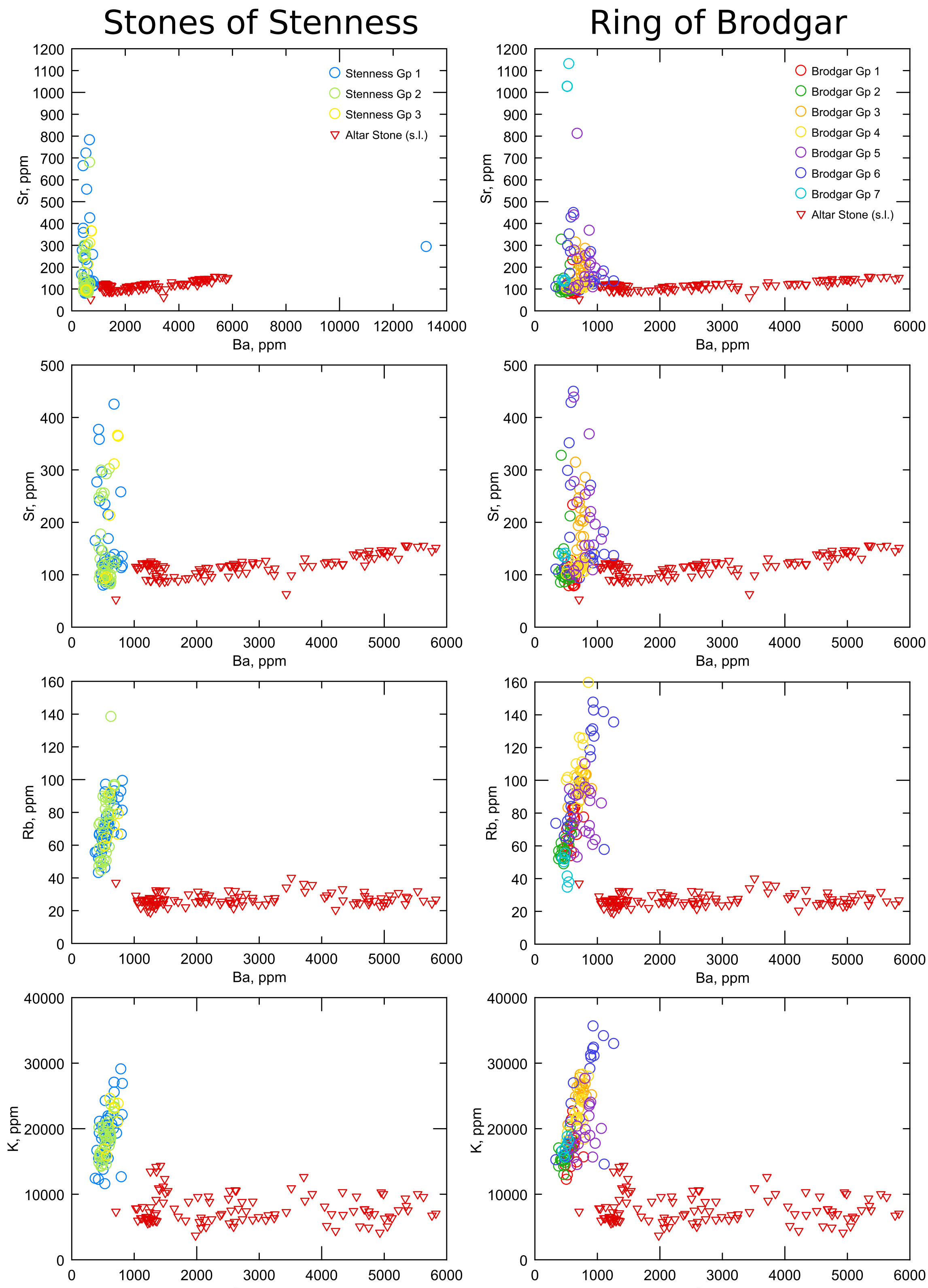

Fig. 8. Bivariate plots for compositional data from the Stonehenge Altar Stone compared with the Stones of Stenness and Ring of Brodgar. Bevins et al. (2023a) used a similar range of elements in attempts to define possible Old Red Sandstone sources in the Anglo-Welsh Basin for the Altar Stone.

This diagram was used by Bevins et al. (2023b) to show the compositional differences between the Stonehenge Altar Stone and ORS lithologies from the Anglo-Welsh Basin. Again, it is clear there is no overlap of the fields of data, and that the Orkney standing stones are compositionally different from the Altar Stone.

Chemical data clearly indicate that the Altar Stone at Stonehenge is not related to the standing stones at either the Ring of Brodgar or the Stones of Stenness. Next, we compare the composition of the Altar Stone

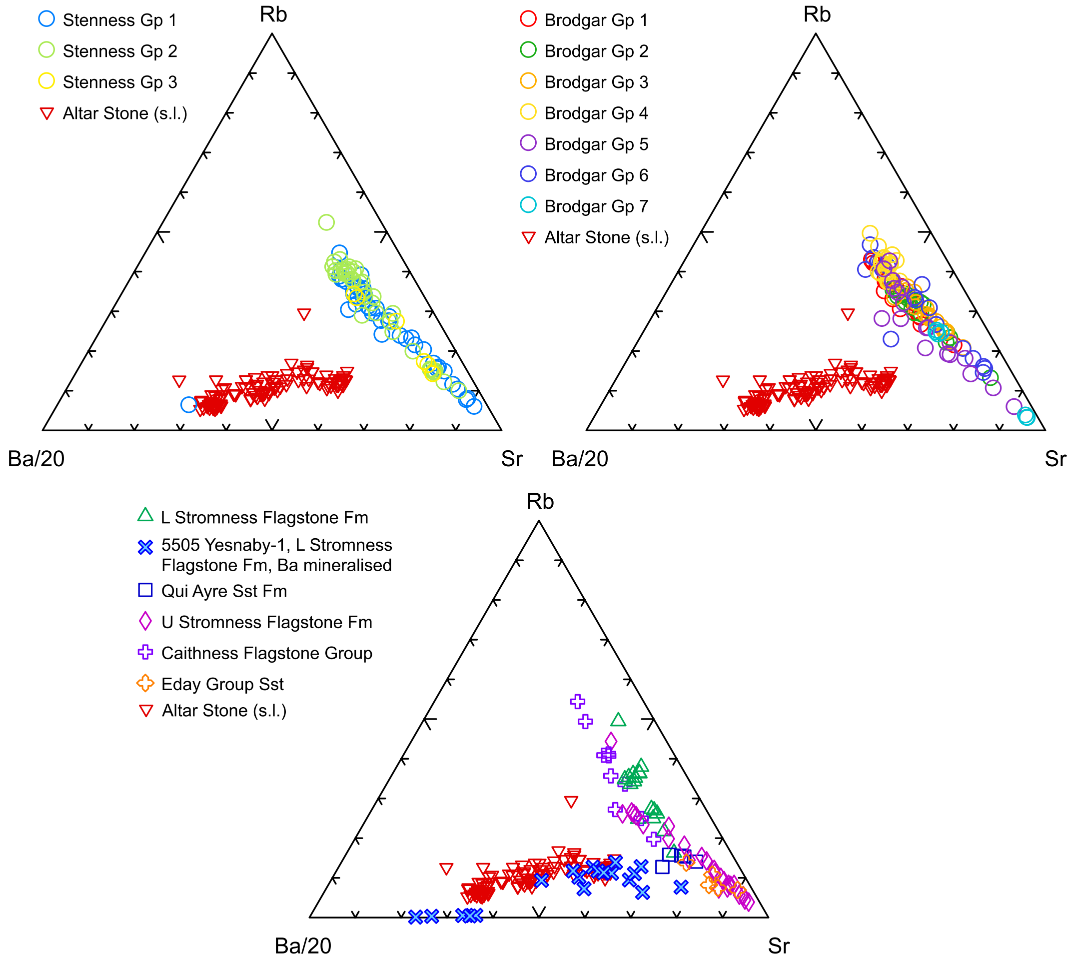

Fig. 9. Triangular diagrams of the Ba-Rb-Sr composition of the Stonehenge Altar Stone compared with data for stones from Stenness and Brodgar.

with local Mainland Orkney ORS lithologies. The Orkney lithologies analysed by pXRF are compared with the Altar Stone in Fig. 9 (Ba-Rb-Sr triangular diagrams), Fig. 10 (Ba vs Sr) and Fig. 11 (Ba vs Rb), where the compositions of the different lithologies are also compared with the analyses of the stones from Stenness and Brodgar, to see if any suggestions as to their provenance can be proposed.

In Fig. 9 the only Orcadian lithology sample analysed which has any similarity to the Altar Stone in Ba-Rb-Sr space is sample 5505 (Yesnaby 1) from the Lower Stromness Flagstone Formation, where there is some compositional overlap. This sample has a mineralised fracture surface along the margins of a thin vein (see Fig. 6), which contained between 9.2–26 wt% Ba, and body compositions in the range 2600–6200 ppm, the latter being similar to the range of Ba displayed by the Altar Stone (see Fig. 9). The Yesnaby 1 sample vein also contains about 3000 ppm Sr, with the body of the rock ranging between 220–700 ppm Sr, but this is between 2–5 x higher than the Altar Stone (Fig. 10) and clearly separates this lithology from the Altar Stone composition.

The baryte mineralisation in sample 5505 (Yesnaby 1) causes compositional mixing between the Lower Stromness Flagstone Formation sandstone and the mineralised vein material (see Fig. 10). From this the Ba/Sr composition of the mineralising fluid can be deduced. This mixing relationship is also seen in Fig. 11 for sample 5505, where Rb, low in the mineralised vein, is reduced in (by leaching/dissolution from) the Lower Stromness Flagstone Formation sandstone as the Ba content increases. This sample attests to localised Ba mineralisation of the ORS on western Mainland, Orkney, as described in Heptinstall et al. (2023) but the Ba/Sr ratio differs markedly from the Altar Stone (see Figs. 9, 10 and 11). If the Altar Stone is a baryte-mineralised lithology from Orkney, then a similar Ba-Sr relationship should exist between a possible unmineralized source and the high-Ba and Sr for the Altar Stone. For the Altar Stone, a positive Ba-Sr relationship exists, and this intersects low Sr samples from both the (undifferentiated) Caithness Flagstone Group from the Upper Stromness Flagstone Formation, notably those analyses from sample 5517, from Skipy Geo (Fig. 10). However, when these lithologies are considered in terms of their Ba and Rb contents (Fig. 11), the Altar Stone shows slightly increasing Rb with increasing Ba, and this ratio does not intersect the compositions of the Orcadian Caithness Flagstone Group analyses, thus ruling out a possible relationship. The Altar Stone Ba-Rb regression also does not intersect the field of Skipy Geo Rb analyses (which are notably higher), again indicating that there is no relationship between these samples. The compositional evidence thus indicates there is no direct link between the Altar Stone and the lithologies analysed from Orkney, whether mineralisation has affected their compositions or not. It is clear from the Ba vs Sr and the Ba vs Rb relationships that the Altar Stone does not resemble chemically any of the lithologies analysed from Orkney.

Finally, whist the geochemical data from the Orcadian lithology analyses show no link with the Altar Stone at Stonehenge, we further explore whether the Orkney lithology compositional data tells us anything of the provenance of the stones used to construct the circles at Brodgar and Stenness Fig. 10 shows the Ba vs Sr compositions of the Orcadian lithologies and the stones from Brodgar and Stenness, and Fig. 11 shows Ba vs Rb. It is clear that there is significant compositional overlap for the Orkney lithologies in terms of these elements, but some general observations can be made. The more quartz-rich sandstones of the Qui Ayre Formation and Eday Group have lower average Rb and Sr

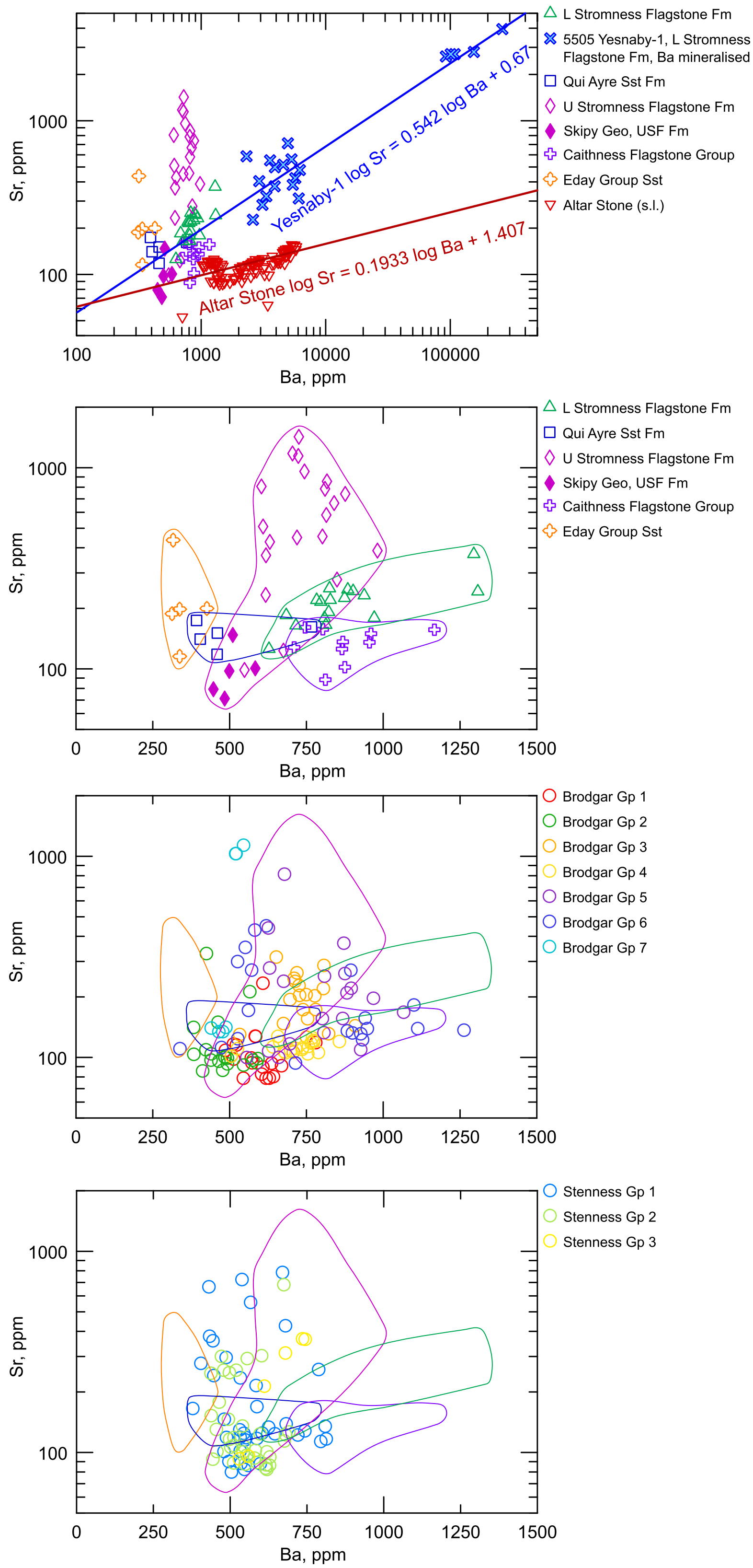

Fig. 10. Comparison of Orkney lithologies (second panel, and field outlines in lower panels) with the Altar Stone, the Stones of Stenness and the Ring of Brodgar in terms of their Ba vs Sr compositions. Note the logarithmic axis for Sr on all panels, and for Ba in the upper panel, which includes the Ba mineralised Lower Stromness Formation sample, Yesnaby-1 (REB 5505): Yesnaby 1 is excluded from the lower plots. See text for discussion.

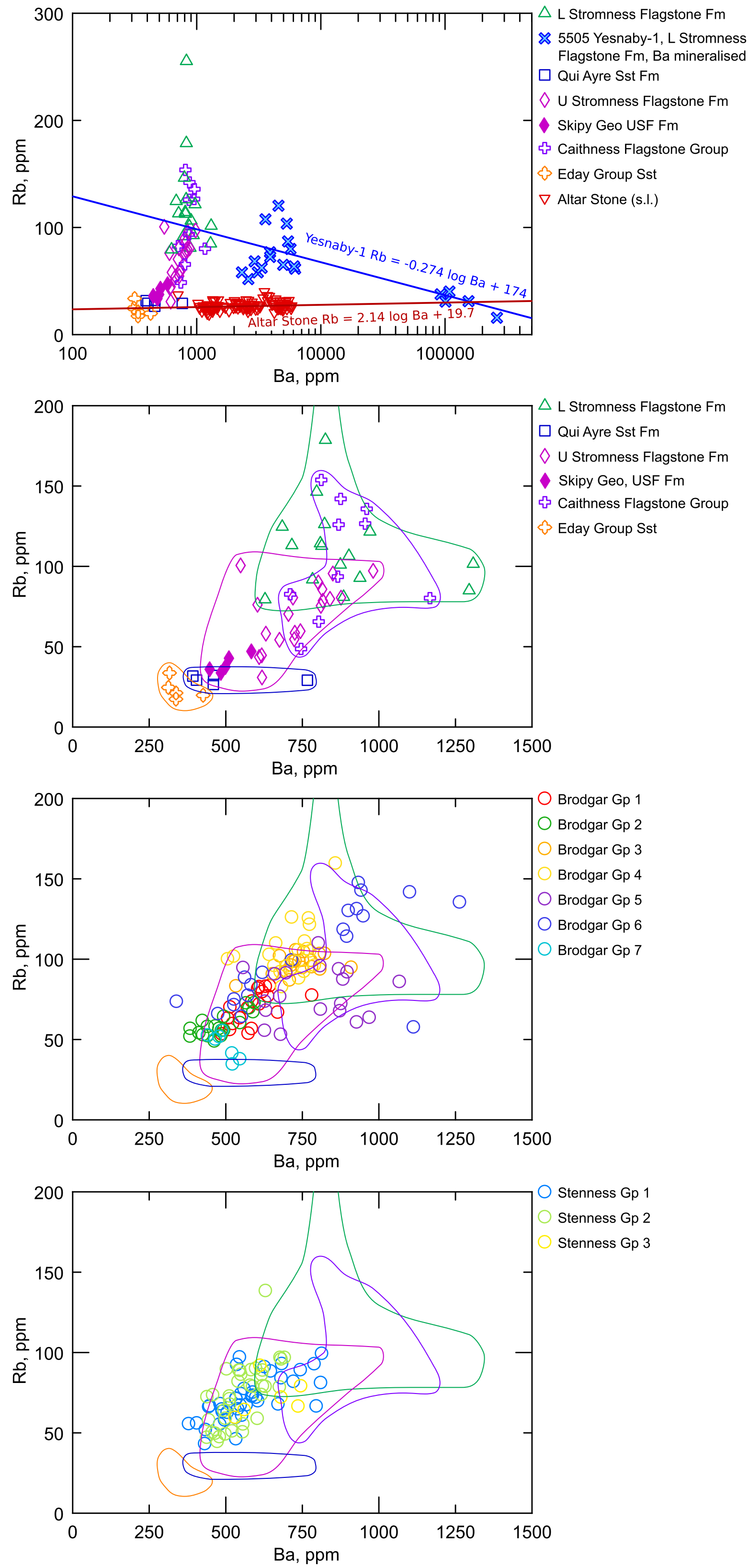

Fig. 11. Comparison of Orkney lithologies (second panel, and field outlines in lower panels) with the Altar Stone, the Stones of Stenness and the Ring of Brodgar in terms of their Ba vs Rb compositions. Note the logarithmic axis for Ba in the upper panel, which includes the Ba mineralised Lower Stromness Formation sample, Yesnaby-1 (REB 5505): Yesnaby 1 is excluded from the lower plots. See text for discussion.

than the Lower and Upper Stromness Flagstone formations and the (undifferentiated) Caithness Flagstone Group.

The Ba vs Sr and Ba vs Rb data both suggest that the analysed Stenness stones are compositionally like the Upper Stromness Flagstone Formation, while the Brodgar stones overlap compositionally with both the Upper and Lower Stromness Flagstone formations, as well as possibly some similarities with the Caithness Flagstone Group (Brodgar Group 6 and possibly Group 5). The Ba vs Rb data suggests that there are

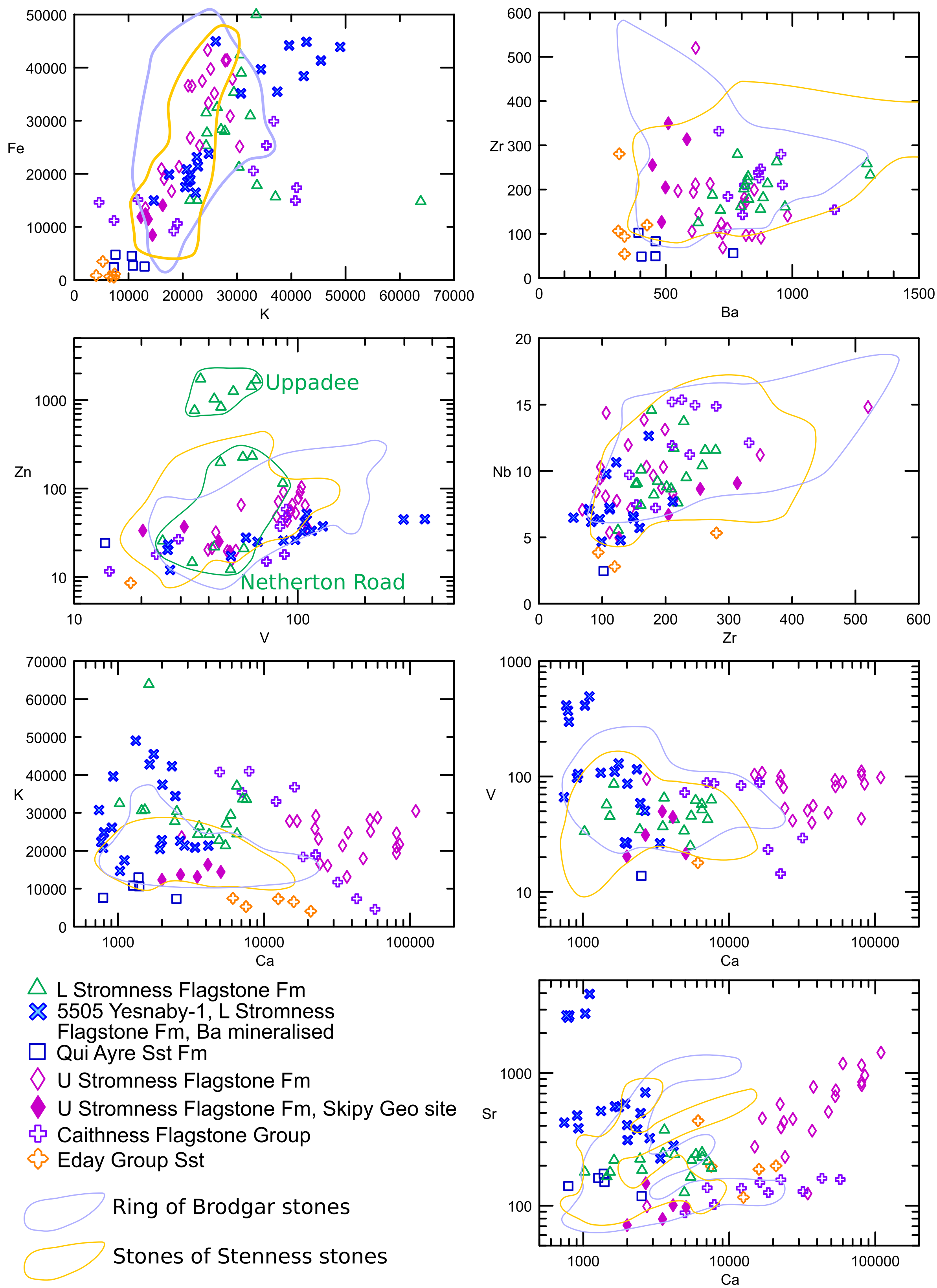

Fig. 12. Selected bivariate plots comparing the compositions of the Stenness and Brodgar stones (compositional fields marked) with the Orcadian lithology analyses.

no stones at Stennes or Brodgar (with the possible exception of Brodgar Group 7) which have been sourced from the Qui Ayre Formation nor from the Eday Group, but these overlap the Lower and Upper Stromness Flagstone formations in terms of their Ba vs Sr compositions. According to Richards (2019) the Group 4 stones at the Ring of Brodgar (which includes Stone 16) derive from a quarry at Staneyhill, in the Upper Stromness Flagstone Formation, but in terms of Ba vs Sr and Ba vs Rb, Stone 16 has more affinity with the Lower Stromness Flagstone Formation (see Figs. 9 and 10). All the stones from Stenness have a composition consistent with the Upper Stromness Flagstone Formation, but Richards (op. cit.) also suggests that none of these is from Staneyhill. Whilst both Upper and Lower Stromness Flagstone formations sandstones crop out either side of the present-day causeway at the Ness of Brodgar, the area around Stenness is dominated by the Upper Stromness Flagstone Formation. The quarry site at Vestra Fiold (Richards et al., 2019) has been suggested as one source for the Brodgar stones, particularly those stones in Groups 1 and 3. This quarry lies about 10 km NNW of the Brodgar circle, sits on Upper Stromness Flagstone Formation sandstones, and the compositions of these groups of Brodgar Stones is consistent with this suggestion. However, the ground between Vestra Fiold and the Ring of Brodgar is underlain by both Lower and Upper Stromness Flagstone formations lithologies (British Geological Survey, 2024), both of which crop out within about 1.5 km of the Ring of Brodgar on the Ness of Brodgar isthmus.

Fig. 12 shows a selection of other bivariate plots of the compositions of Orcadian lithologies, with fields showing the compositions of stones analysed from Stenness or Brodgar. As with Ba vs Sr or Ba vs Rb, the Stenness stones show compositions which overlap with the Upper Stromness Flagstone for many elements (e.g. K vs Fe, Ba vs Zr, Zr vs Nb), whereas the Brodgar stones show a wider compositional range, encompassing both Lower and Upper Stromness Flagstone Formation compositions. Again, the standing stones' compositions do not overlap the compositional fields of the Eday Group or Qui Ayre Sandstone Formation (notably K vs Fe), and in these two formations many elements are below the lower limits of detection (LoD), reflecting the highly siliceous nature of these sandstones. In addition to the Ba mineralisation noted above in sample 5505 (Yesnaby 1) of the Lower Stromness Flagstone Formation, Zn mineralisation is also evident in this unit at Burn of Uppadee, where Zn contents reach around 2000 ppm caused by Cu-U-REE mineralisation related to oil emplacement (Heptinstall et al., 2023). Fig. 13 shows Ba vs U data for the Brodgar stones, where one stone, Stone 15 (Group 6) shows an unusually and consistently high U, at around 20 ppm, about twice the normal maximum U concentrations recorded in the other lithologies analysed (many of which are below the LoD of about 5 ppm), and this may either relate to the redox conditions during sediment deposition, or may be related to the same phase of Cu-U-REE mineralisation. Whatever the cause, this may provide a chemical indicator which could help provenance the source of this orthostat.

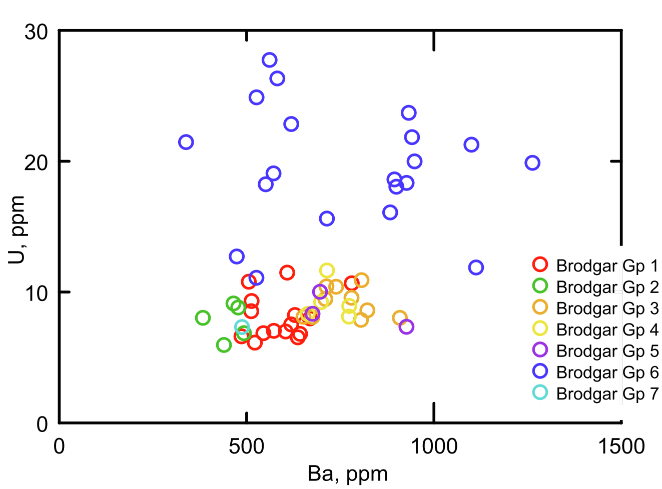

Fig. 13. Concentrations of Ba vs U from stones from the Ring of Brodgar. Detection limit for U by pXRF is about 5 ppm, and the majority of analyses fall below this.

Overall, trace element compositions show similarities between the Stenness and Brodgar stones and the Lower and Upper Stromness Flagstone formations (Figs. 9, 10 and 11), as do the less mobile major elements (e.g. Fe and K). However, Ca, present at concentrations up to ~ 10 wt% in the Upper Stromness Flagstone Formation, largely as the mineral calcite, generally exceeds standing stone compositions, which are all ~ 2 wt% (see Fig. 12). The low Ca in the standing stones will be related to protracted leaching of calcite by exposure to the elements since their erection, and similar observations have been made for the surface of the Stonehenge Altar Stone and its debitage fragments (Bevins et al., 2022a; Bevins et al., 2023b). The Lower Stromness Flagstone Formation does not contain more than ~ 1 wt% Ca and has low calcite contents and generally higher dolomite/ankerite which readily accounts for all the Ca in the analyses. Downes et al. (2019) suggested that fallen stone F4 from Brodgar (Group 7) was sourced from the Eday Group – the low non-SiO_2 major and trace element compositions in this stone (Figs. 10 and 11) suggest a pure sandstone, but the Ba, K, and Rb data suggest the Qui Ayre Sandstone is also a possibility and is compositionally closer to F4. Both of these lithologies have > 80 modal% quartz (determined by XRD), in contrast to the remaining sandstones analyses which have < 57 modal% quartz.

6. Discussion

The compositional data for the standing stones at Brodgar and Stenness is similar to that from the Lower and Upper Stromness Flagstone formations while the Stones of Stenness appear (on chemical grounds) to have come from the Upper Stromness Flagstone Formation and those from Brodgar from both units. One Brodgar sample (F4, Group 7) may be sourced from the Qui Ayre Sandstone Formation or possibly the Eday Group, whilst another contains high U (Brodgar stone 15, Group 6) which may be helpful in proving its source in future. For the most part however, the chemistry of the Stenness and Brodgar stones offer no particular characteristics which would be helpful in identifying their provenance. In all cases, comparison with the Altar Stone at Stonehenge shows no compositional overlap with the Stenness and Brodgar stones, nor any of the Orcadian lithologies analysed, and it can be concluded that the Altar Stone has no direct link to these Orcadian sites.

A key comparative feature from the mineralogical analyses is the presence of relatively abundant K-feldspar in all of the Orkney field samples, which contrasts with only trace abundance of K-feldspar in the Altar Stone (Fig. 14). Clay mineral assemblages of the field samples from Orkney are also not comparable with the clay mineral assemblage in the Altar Stone, in particular the presence of tosudite (dioctahedral chlorite/smectite) in the Altar Stone which is a key feature that is not matched in the Orkney samples. As previously shown by Hillier and Clayton (1989) the clay mineral assemblage of large parts of the flagstone sequences in both Orkney and Caithness are often solely composed of illite. Whilst in other parts, assemblages consisting of illite, trioctahedral chlorite plus or minus chlorite/smectite or corrensite (both trioctahedral) are common. Although the publications of Hillier and Clayton (1989) and Hillier (1993) were focused on mudstones and fine grained lithologies, it was also noted in the thesis of Hillier (1989) that the associated sandstones tended to mirror the mudstones in terms of their clay mineral assemblages. However, two samples in the present set were somewhat different. A sample from the Eday Group (5514) showed a clay mineral assemblage similar to that seen in other parts of the Orcadian Basin, such as around the Cromarty Firth, consisting mainly of an R1 ordered mixed-layer illite/smectite with an expandability of around 25 %, together with abundant kaolinite (see Table 4). Note that kaolinite is also present in the Altar Stone. The other sample showing a marked difference to

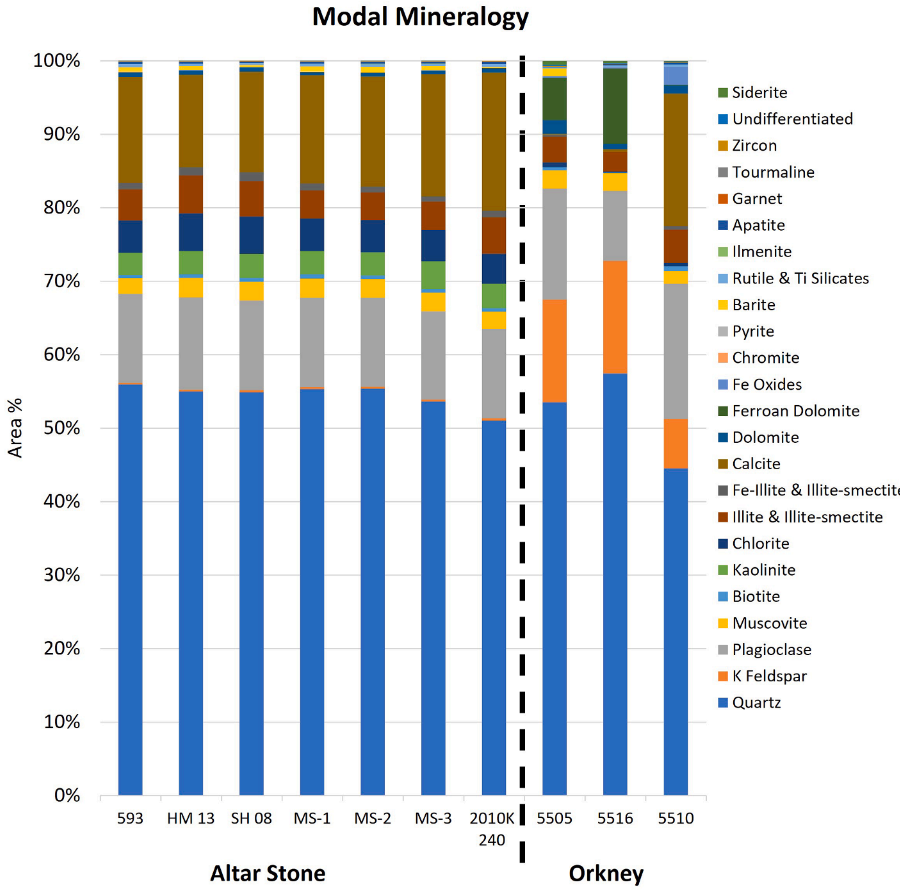

Fig. 14. Comparison of the modal mineralogy as determined through automated SEM-EDS analysis for (a) samples derived from the Altar Stone, Stonehenge vs (b) three sandstone samples from Orkney. Samples 5505 and 5516 from the Lower Stromness Flagstone Formation. Sample 5510 from the Upper Stromness Flagstone Formation.

other Orkney samples in the present set is sample 5512, from the (undifferentiated) Caithness Flagstone Group, which contains a moderate amount of a dioctahedral smectite. Given that the bulk rock XRD analyses of the samples also showed the presence of goethite, which is presumably an indication of surface weathering of the samples (ochre colours forming thin films of iron oxides are often seen at outcrops and no attempt was made to avoid such parts of the samples during preparation) we assume that the smectite may be a modern weathering product. In any case smectite is not present in the clay mineral assemblage of the Altar Stone.

As has been shown by Bevins et al. (2022a, 2023b), baryte is present as a cement in the Altar Stone. Baryte was observed in two of the analysed Orkney ORS samples, namely 5507 from the Qui Ayre Sandstone Formation at Yesnaby and sample 5505 from the Lower Stromness Flagstone Formation, also from Yesnaby. The baryte in these two samples from the Yesnaby area appears to be related to mineralization and is, at least in part, occurring as vein infills (Fig. 6), which is not the case for the Altar Stone baryte. The overall mineralogical profile for both the Qui Ayre sample and the Lower Stromness Flagstone Formation, clearly shows that the Altar Stone could not have been derived from these sources.

7. Conclusions

Portable XRF analyses of the five exposed stones at the Stones of Stenness and seven of the exposed stones at the Ring of Brodgar show a wide range of compositions, covering similar compositional spaces on geochemical plots as field samples analysed from both the Lower and Upper Stromness Flagstone formations, with the stones at Stenness appearing to have been sourced from the Upper Stromness Flagstone Formation while the Ring of Brodgar stones possibly being sourced from both formations. Geochemical data suggest that one Ring of Brodgar stone (F4, Group 7) may have been sourced from either the Qui Ayre Sandstone Formation or the Eday Group, the latter horizon having been proposed as the source of stone F4 by Downes et al. (2019). What is clear, however, is that the chemistry of the Stenness and Brodgar stones obtained by pXRF do not have any distinctive characteristics which are of value in terms of identifying their source.

Examination of the mineralogy of ORS field samples and the Stonehenge Altar Stone using a combination of X-ray diffraction, microscopy, Raman spectroscopy and automated SEM-EDS shows there to be no match between the Orkney samples and the Altar Stone. Only two samples from Orkney showed the presence of baryte, a characteristic mineral of the Altar Stone, one showing just a trace amount not detected in the HM analysis; both appear to be related to mineralization, which is not the case for the Altar Stone baryte. Another key discriminant is the presence of abundant detrital K-feldspar in all of the Orkney field samples, a mineral which has only very low abundance in the Altar Stone. In addition, the regularly interstratified dioctahedral chlorite/dioctahedral smectite mineral tosudite is present in the clay mineral assemblage of the Altar Stone, but not detected in the Orkney samples.

The conclusion reached here, based on the mineralogical and geochemical investigations undertaken, is that despite Clarke et al. (2024) identifying an Orcadian Basin source for the Stonehenge Altar Stone, it was not sourced from Mainland Orkney, notwithstanding the abundant evidence for long distance connections between Stonehenge and Orkney around 3000/2900 BCE, as detailed in the introduction to this paper. The mineralogical and compositional evidence presented here will assist in identification of possible source lithologies for the Altar Stone in the future.

CRediT authorship contribution statement

Richard E. Bevins: Conceptualization, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Writing – original draft, Writing – review & editing. **Nick J.G. Pearce:** Conceptualization, Formal analysis, Investigation, Methodology, Software, Writing – original draft, Writing – review & editing, Visualization. **Stephen Hillier:** Formal analysis, Investigation, Methodology, Software, Writing – original draft. **Duncan Pirrie:** Formal analysis, Investigation, Methodology, Software, Writing – original draft. **Rob A. Ixer:** Formal analysis, Investigation, Writing – review & editing. **Sergio Andò:** Formal analysis, Investigation, Visualization, Writing – original draft, Writing – review & editing. **Marta Barbarano:** Formal analysis, Methodology, Writing – review & editing. **Matthew Power:** Formal analysis, Methodology, Software, Writing – review & editing. **Peter Turner:** Investigation, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data submitted as Supplementary tables

Acknowledgements

Professor Colin Richards (University of Highlands and Islands) is thanked for fruitful discussions at the early stage of this project. Dr Aurélie Turmel and Dr Lisa Brown (both Historic Environment Scotland) are thanked for assisting with necessary permissions. Richard Bevins gratefully acknowledges receipt of an Emeritus Fellowship from the Leverhulme Trust.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jasrep.2024.104738.

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