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# TECTONOSTRATIGRAPHIC AND PALEOENVIRONMENTAL SETTINGS OF HOST-REPLACING PHREATIC CALCRETE HARDPANS DEVELOPED AT BASIN MARGINS IN THE UPPER DEVONIAN KINNESSWOOD FORMATION OF SOUTHWEST SCOTLAND

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| Abstract: | In the Firth of Clyde area of southwest Scotland, the Famennian Kinnesswood Formation includes an interval of massive, host-replacing phreatic calcrete hardpan (HRPCH), the likes of which have only been documented at few locations and few intervals in geological history. The HRPCH is only found at basin margin shoulders, where the Kinnesswood Formation succession is thin and incomplete. The isles of Bute and Great Cumbrae provide well exposed sections in which adjacent shoulder and trough successions can be correlated and compared to clarify the tectonostratigraphic and paleoenvironmental settings of the HRPCHs. In the Cumbraes Trough, above a thin interval of peritidal limestone, the middle part of the Kinnesswood Formation (lower part of the Foul Port Member) is pervasively disturbed by large syn-depositional dewatering structures interpreted to be products of the intermittent deposition and dissolution of evaporites. These structures occur at approximately the same stratigraphic interval as the HRPCHs on the isles of Bute and possibly Arran. The HRPCH in Bute is interpreted to have developed on a syn-depositional shoulder adjacent to a growth fault (the Kerrycroy Fault) delimiting a trough that accommodated intermittent sea water incursions in a restricted, evaporitic setting. This is consistent with the current model for HRPCH formation, which involves the mixing of fresh groundwater issued from source areas with the high pH groundwater that surrounds evaporitic basins. A significant increase in silica solubility paired with a decrease in calcite solubility occurs in the mixing zone, thus promoting the thorough replacement of silicates with calcrete. |

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**ABSTRACT: In the Firth of Clyde area of southwest Scotland, the Famennian Kinnesswood Formation includes an interval of massive, host-replacing phreatic calcrete hardpan (HRPCH), the likes of which have only been documented at few locations and few intervals in geological history. The HRPCH is only found at basin margin shoulders, where the Kinnesswood Formation succession is thin and incomplete. The isles of Bute and Great Cumbrae provide well exposed sections in which adjacent shoulder and trough successions can be correlated and compared to clarify the tectonostratigraphic and paleoenvironmental settings of the HRPCHs. In the Cumbraes Trough, above a thin interval of peritidal limestone, the middle part of the Kinnesswood Formation (lower part of the Foul Port Member) is pervasively disturbed by large syn-depositional dewatering structures interpreted to be products of the intermittent deposition and dissolution of evaporites. These structures occur at approximately the same stratigraphic interval as the HRPCHs on the isles of Bute and possibly Arran. The HRPCH in Bute is interpreted to have developed on a syn-depositional shoulder adjacent to a growth fault (the Kerrycroy Fault) delimiting a trough that accommodated intermittent sea water incursions in a restricted, evaporitic setting. This is consistent with the current model for HRPCH formation, which involves the mixing of fresh groundwater issued from source areas with the high pH groundwater that surrounds evaporitic basins. A significant increase in silica solubility paired with a decrease in calcite solubility occurs in the mixing zone, thus promoting the thorough replacement of silicates with calcrete.**

# INTRODUCTION

Erosional remnants of massive, host-replacing phreatic calcrete hardpans (HRPCHs) were documented at basin margins in the lower part of the Upper Devonian to Lower Carboniferous Inverclyde Group on the isles of Arran (Jutras et al. 2011) and Bute (Young and Caldwell 2011a) in southwest Scotland (Fig. 1). Whereas 1-30 cm thick lenses of invasive phreatic calcrete developed by mineral displacement in porous sediment between muddy aquicludes are relatively common (e.g., Tandon and Friend 1989; Jutras et al. 2001, 2016; Hillier et al. 2011), evidence for thorough mineral replacement by phreatic calcrete over a thickness exceeding 1 m is notably rare in the geological record. The latter has only been reported from the Upper Devonian to Lower Carboniferous successions of eastern Canada and southwest Scotland (Jutras et al. 1999, 2001, 2007a, b, 2011; Jutras and Prichonnet 2002, 2005; Young and Caldwell 2011a; Jutras, 2017), and from Quaternary deposits of Western and Central Australia (Butt et al. 1977; Mann and Horwitz 1979; Arakel and McConchie 1982; Jacobson et al. 1988; Arakel et al. 1989). Based on the model developed by Arakel and McConchie (1982) for modern occurrences, such thick (1-12 m) and massive HRPCHs are usually assumed to form in hyper-arid environments where calcium-bearing fresh groundwater flows into and mixes with the high pH groundwater of an evaporitic aquifer (e.g., Jutras et al. 2011; Jutras 2017), which causes an increase in silicate solubility paired with a decrease in calcite solubility, and which results in a thorough replacement of the former with the latter. However, this relationship between HRPCH and adjacent evaporites was only demonstrated in a few studies (e.g., Arakel and McConchie 1982; Jutras et al. 2007b).

In the Inverclyde Group of southwest Scotland, the HRPCH intervals only occur at basin margin shoulders and do not occur in adjacent troughs. In this paper, well exposed sections of the

Inverclyde Group on the Bute Shoulder and in the adjacent Cumbraes Trough are compared using field data and the stable isotopic analysis of carbonate samples in order to determine the tectonostratigraphic and paleoenvironmental settings of the HRPCH occurrences, as well as to improve our general understanding of the petrogenetic setting of this rare type of calcrete.

## GEOLOGICAL SETTING

The Upper Devonian to Lower Carboniferous successions of Scotland were deposited in post-orogenic, fault-bounded successor basins (Read et al. 2002) in response to a nearly N-S (~355°-175 °) shortening direction (Coward 1993). The Inverclyde Group is comprised of three formations that can be correlated across the Midland Valley of Scotland (Read et al. 2002). The basal Kinnesswood Formation is mostly characterized by red clastic rocks with abundant calcretes, the conformably overlying Ballagan Formation by gray mudrock and sandstone, and the top Clyde Sandstone Formation by thick bodies of coarse gray sandstone with intervals of red or purple mudrock (Paterson and Hall 1986). In the Firth of Clyde area of southwest Scotland (Fig. 1), these units show significant differences in thickness (by over one order of magnitude) on closely spaced fault blocks (Young and Caldwell 2011a, 2012). Whereas the contact between the Kinnesswood Formation and the underlying Famennian Upper Old Red Sandstone (UORS) is generally interpreted as conformable in the Midland Valley (Read et al. 2002), it is disconformable on the Isle of Arran (Jutras et al. 2011; Young and Caldwell 2012). Until recently, the UORS/Kinnesswood contact was believed to approximately correspond to the Devonian-Carboniferous boundary (e.g., Read et al. 2002). However, recent work from Marshall et al. (2019) suggests that the Kinnesswood Formation is Famennian and that the Carboniferous succession in Scotland starts with gray beds of the Tournaisian Ballagan Formation. The Clyde Sandstone Formation is undated, but well constrained between the Courceyan to early Chadian

Ballagan Formation (Marshall et al. 2019) and the early Asbian Clyde Plateau Volcanic Formation (CPVF; Pointon et al. 2014) (Fig. 2).

## METHODOLOGY

The best exposed sections of the Upper Devonian to Lower Carboniferous successions on the isles of Bute and Great Cumbrae were measured and studied in terms of their petrography, sedimentology, stratigraphy, and eodiagenetic features, with emphasis on calcretes. Paleocurrent vectors were obtained from both successions to determine the approximate paleo-position of source areas (Supplementary Appendix 1). In relatively coarse and proximal successions, such as those in this study, source areas are best determined with trough channel orientations, which mark times of high energy when currents are least deviated by sediment engorgement (see Jutras et al. 2015 for a discussion). In Bute, trough channel orientations were measured at Dunagoil Bay with additional data from a section fragmented by faults and dykes at Hawk's Neb on the southeast coast (Fig. 1). In Great Cumbrae, paleocurrent measurements were obtained from the southwest coast and Foul Port areas (Fig. 1).

The study also includes analyses of stable carbon and oxygen isotopes ($\delta^{13}C$ VPDB and $\delta^{18}O$ VPDB) from a total of 22 samples of 95-100% pure carbonate rocks (intrasparudite as well as micritic material from various types of calcrete) in the Inverclyde Group on both isles. None of the sampled areas have been affected by post-formational veining, and all calcrete samples are from the center of massive nodules and hardpans, which are impermeable features that mostly act as closed systems during deep burial diagenesis. The samples were powdered and analyzed by dual inlet mass spectrometry at the GEOTOP lab of Université du Québec à Montréal

(Canada) using a GV Instruments Multicarb preparation system connected to an Isoprime Dual Inlet mass spectrometer (Supplementary Appendix 2).

## UPPER DEVONIAN TO LOWER CARBONIFEROUS STRATIGRAPHY IN GREAT CUMBRAE

The thick Upper Devonian to Lower Carboniferous succession in Great Cumbrae is incomplete and partly obscured by numerous dykes and faults. However, large portions of it are well exposed. On the southwest coast, the upper part of the UORS is comprised of thick, cross-cutting channels of orange-red, poorly-sorted and polymictic gravelly sandstone devoid of calcretes (Fig. 3). The UORS is separated from the Kinnesswood Formation by a large felsic dyke. Strike and dip of the two units is different, suggesting the contact is a low-angle unconformity (Fig. 3).

### *The Kinnesswood Formation Doughend Sandstone Member*

Basal beds of the Kinnesswood Formation on the southwest coast of Great Cumbrae were assigned to the Doughend Sandstone Member by Monro (1999). Compared with the underlying UORS, this unit is characterized by thinner, wider and better sorted channels of sandstone, mudstone and conglomerate, the latter being considerably more mature in terms of clast composition (~90% quartz pebbles versus ~10% in the underlying UORS). The Doughend Sandstone Member is also much more calcitic, which gives it a pinkish-red colour, and it includes thin lenses of phreatic calcrete as well as some calcrete nodules, which are absent in the underlying UORS (Fig. 3). The upper part of the Doughend Sandstone Member is obscured by exposure gaps and a series of large mafic dykes (Fig. 3).

### *The Kinnesswood Formation Foul Port Member*

Directly above the uppermost dyke that truncates the upper part of the Doughend Sandstone Member, a ~1 m thick exposure of brownish-red mudrock with calcrete nodules transitions upward into a few centimetres of gray marl that is sharply overlain by a well-bedded, ~0.75 m thick intrasparudite (Fig. 4A). The calcarenitic top of this limestone interval is characterized by well-developed interference ripple marks that are enhanced by dewatering (Fig. 4B). Such features are typical of peritidal carbonates (Wright 1984; Pratt 2010). There is a large exposure gap above this thin limestone interval on the southwest coast, but sparse outcrops suggest that the overlying succession is mainly characterized by brownish-red mudstone that is similar to the one that is directly underlying it. This brownish-red mudrock succession forms the bulk of the Kinnesswood Formation Foul Port Member, which near its base includes the above-mentioned peritidal carbonate, referred to as the Doughend Limestone marker (Paterson and Hall 1986; Monro 1999; Young and Caldwell 2011b).

Although incomplete and partly obscured by faulting, the Foul Port Member is best exposed in its type-area of Foul Port on a wave-cut platform developed along the south coast of Great Cumbrae (Figs. 1 and 3). Considering the lack of stratigraphic repetition across faults, a minimum thickness of ~200 m can be estimated for the Foul Port Member in Great Cumbrae. Based on extrapolations from mapping data, Caldwell and Young (2013a) consider this member to be ~280 m thick in Great Cumbrae. Although brownish-red mudrock dominates the entire unit, minor sandstone and gravelly sandstone intervals occur throughout the Foul Port Member, but are significantly more abundant in its upper half (Fig. 3). The mudstone is mottled by pedogenic calcite concretions in many intervals, but well-developed calcrete nodules only occur in the upper half of this member (Fig. 3).

**Large dewatering structures at Foul Port.—**In the basal ~47 m exposure of mostly mudstone at Foul Port, most intervals are pervasively disturbed by large tepee-like structures that are ~0.5 m tall on average and separated by synclinal structures that are ~1 m wide on average (Figs. 3 and 5). The synclinal structures are areas cemented by pedogenic calcite, which is mostly absent from the brecciated "tepee" structures (Fig. 5). These structures are interpreted to have developed at intervals where dewatering was obstructed by calcrete cement, thus forcing its concentration in discrete escape pathways. Because they are clearly associated with volume loss, the synclinal structures are herein referred to as "pseudo-synclines" to differentiate them from what is often referred to in the literature as "pseudo-anticlines", which are less closely spaced structures that are either associated to volume gain from an excessive accumulation of calcrete or dolocrete cement (e.g., Price 1925; Jennings and Sweeting 1961; Watts 1977; Botha and Hugues 1992; Eren 2007; Francis et al. 2007) or from the re-watering of pedogenic clays in confined settings (e.g., Driese and Foreman 1992; Caudill et al. 1996; Driese et al. 2000).

At some intervals of the same basal succession at Foul Port, dewatering structures are larger and characterized by isolated blocks of calcretized mudrock that exhibit evidence of folding, rotation and sinking, again separated by brecciated mudrock in which pedogenic calcite is mostly absent (Figs. 3 and 6). These larger structures are interpreted to reflect dewatering from thicker intervals of wet clays.

Other intervals exhibit wavy soft-sediment deformations enhanced by fibrous calcite (Figs. 3 and 7), referred to a "scalloped calcite veins" by Caldwell and Young (2013a). Pedogenic calcretic concretions are absent in these intervals. These soft-sediment deformations are interpreted to have occurred at intervals where dewatering was not significantly obstructed by calcrete cement.

All three types of dewatering structures are sharply truncated in some intervals by a thin sheet of sandstone (Figs. 3 and 7). These truncations indicate that the associated dewatering structures formed in very shallow burial conditions, shortly after deposition, and prior to deposition of the directly overlying beds. Each of these sparse and thin sandstone sheets include granules of calcretic concretions and often bear ripple marks at their top. They form less than 1% of the succession and are interpreted as the record of occasional sheetfloods in the succession.

Whereas pseudo-syncline-tepee assemblages and isolated blocks between brecciated dewatering pathways are only observed in the basal ~47 m at Foul Port, mudstone intervals with wavy calcite extend significantly higher in the succession (Fig. 3). However, the uppermost ~55 m interval of the Foul Port Member is mostly devoid of dewatering structures (Fig. 3).

### *The Ballagan and Clyde Sandstone Formations*

The Kinnesswood Formation at Foul Port is disconformably overlain by the Ballagan Formation, which is dominated by gray sandstone and mudstone intervals that are devoid of calcrete, but which also includes some thin red, purple and green muddy intervals with well-developed nodular calcretes (Fig. 3). One interval includes phreatic calcrete lenses developed in thin sandstone beds between muddy aquicludes (Figs. 3 and 8). The invasive nature of these calcrete lenses is clearly indicated by the pseudo-anticline that they form in the succession (Fig. 8), which suggests mineral displacement rather than replacement.

With uncertainties related to unknown offsets along faults a minimum thickness of ~25 m can be estimated for the Ballagan Formation, which is conformably overlain by the Clyde Sandstone Formation. The latter is incomplete in Great Cumbrae, but is estimated to have a minimum thickness of ~60 m based on tentative extrapolations across faults (Fig. 3). This succession of the Clyde Sandstone Formation includes several thick bodies of gray pebbly sandstone that are similar to the one at the base of the Ballagan Formation (Fig. 3). The Clyde Sandstone Formation is however dominated by thinner and finer purple sandstone, as well as purple mudstone with abundant calcrete nodules. The purple intervals also include some phreatic calcrete lenses developed in sandy intervals between muddy aquicludes (Fig. 3).

## UPPER DEVONIAN TO LOWER CARBONIFEROUS STRATIGRAPHY IN SOUTH BUTE

In Bute, the Kinnesswood Formation is best exposed along a wave-cut platform at Dunagoil Bay in the southwest region of the isle (Young and Caldwell 2011a) (Fig. 1). In this section, the Kinnesswood Formation is unconformably underlain by a thick succession of the UORS, which is similar to that in Great Cumbrae, except for better sorting, with polymictic gravels being concentrated in small channels among larger channels of well-sorted sands (Fig. 9). The low-angle unconformity that separates it from the Kinnesswood Formation is well-exposed (Fig. 10A), although partly masked by syn-Kinnesswood calcrete extending down into the first few centimetres below the unconformity (Fig. 10B). The unconformity is also clearly visible on satellite photos (Fig. 10C).

### *The Kinnesswood Formation Doughend Sandstone Member*

Similar to the succession in Great Cumbrae, the base of the Kinnesswood Formation in Bute is characterized by thin and broad channels of calcitic, pinkish sandstone and quartzose conglomerate that are typical of the Doughend Sandstone Member, with well-developed calcretes in the muddy intervals (Fig. 9). However, the Doughend Sandstone Member is generally coarser in Bute than in Great Cumbrae, suggesting greater proximity to its source area.

Moreover, conglomerates of this unit in Bute are polymictic in the uppermost ~10 m of the succession (Fig. 9), which are unexposed in Great Cumbrae (Fig. 3).

### *Massive Host-Replacing Phreatic Calcrete Hardpan Unit*

The uppermost part of the Kinnesswood Formation at Dunagoil is characterized by ~3.5 m of gray polymictic conglomerate and sandstone invaded by well-developed, ~10 to 20 cm thick phreatic calcrete bands, and sharply overlain by the ~1 m thick erosional remnant of a massive calcrete (Fig. 9). The (1) sharp base, (2) tabular shape, (3) significant thickness, (4) *alpha* (i.e. massive) microfabric (*sensu* Wright 1990), (5) near absence of siliciclastic grains (Fig. 11), and (6) lack of oxidation features (suggesting development below the water table) that characterize the latter calcrete are typical of host-replacing phreatic calcrete hardpans (HRPCHs) (Jutras et al. 2007b, and references therein). Between the bands of phreatic calcrete that underlie the HRPCH, the sandstones and conglomerates are thoroughly invaded by calcrete cement except for some thin intervals of pinkish-red sandstone that were spared from thorough calcretization. Because of their dominantly gray colour, these beds were assigned by Young and Caldwell (2011a) to the base of the Ballagan Formation, but assignment to the uppermost part of the Doughend Sandstone Member is herein proposed in the discussion.

### *The Ballagan and Clyde Sandstone Formations*

A ~15 m gap separates the incomplete HRPCH exposure from a ~1.3 m thick pale gray sandstone overlain by basalt of the Clyde Plateau Formation (Fig. 9). According to Young and Caldwell (2011a), the no longer exposed upper half of this ~15 m interval is dominated by gray mudstones that are typical of the Ballagan Formation. These authors assigned the thin interval of sandstone below the basalt to the Clyde Sandstone Formation (Fig. 9).

## PALEOCURRENTS AND PROVENANCE

In Bute and Great Cumbrae, trough-channel orientations in the UORS and the Inverclyde Group point towards a sediment source area to the southwest (Fig. 1). This is consistent with provenance data, as the petromictic clast contents in these units on both isles are mostly characterized by gray to buff metasedimentary rocks, which are typical of the Eoproterozoic to Lower Paleozoic Dalradian Supergroup that forms the bulk of pre-Late Devonian basement rocks to the southwest (Isle of Arran and Kintyre Peninsula; British Geological Survey 1987, 1996).Conglomerates of the UORS and the Inverclyde Group also include minor occurrences of red sandstone pebbles, which are interpreted to have been mostly sourced from the Lower Devonian Lower Old Red Sandstone, as remnants of the latter unit are also found southwest of the study area (British Geological Survey 1987, 1996). However, the low angle unconformity that separates the UORS from the overlying Kinnesswood Formation implies that some red sandstone pebbles in basal beds of the latter unit were possibly sourced from the former. The data also suggest that the successions in Bute and Great Cumbrae were part of the same basin. This is consistent with the presence of slightly coarser beds in the Doughend Sandstone Member in Bute than in Great Cumbrae, which, based on its current position would have been located farther away from the inferred source area to the southwest (Fig. 1).

Also lying to the southwest are the Northeast Arran Trough and Corrie Shoulder (Fig. 1), which are characterized by very contrasting Lower Carboniferous stratigraphic successions (Jutras et al. 2011; Young and Caldwell 2012), suggesting they were not part of the same basin. If this is the case, it implies that significant post-Early Carboniferous lateral displacement occurred along the large NW-SE trending faults that cut the Firth of Clyde area, as suggested by Young and Caldwell (2012).

## STABLE ISOTOPES OF CARBON AND OXYGEN IN LIMESTONE AND CALCRETES OF THE INVERCLYDE GROUP

The stable isotopic compositions of calcretes in the studied successions are consistent with values from other calcretes of that age in Scotland and eastern Canada (Jutras et al., 1999, 2006, 2007b, 2011; Jutras, 2017; Fig. 12A), which were juxtaposed areas at the time (Scotese and Wright, 2018). However, calcretes from the stratigraphic interval corresponding to the lower half of the Foul Port Member (including the HRPCH at Dunagoil, which is interpreted as belonging the same interval; see discussion) are characterized by heavier $\delta^{13}C$ values than calcretes from intervals that are either above or below (Fig. 12B). Hence, this deviation from the general trend cannot be explained by relative burial depth. Moreover, the deviation towards heavier $\delta^{13}C$ values in the lower Foul Port Member is not co-varying with $\delta^{18}O$ values (Fig. 12B) and therefore cannot be explained by a simple aridification of the climate (*sensu* Stiller et al. 1985; Cerling 1991; Rossinsky and Swart 1993; Andrews et al. 1998; Hsieh et al. 1998; Pentecost 2005; Jutras 2017).

Near the base of the lower Foul Port Member, the peritidal Doughend Limestone marker shows $\delta^{13}C$ values that lie in the higher range of Late Devonian to Early Carboniferous calcretes, just below the recorded range of marine carbonates of that age, which does not overlap with that of calcretes (Fig. 12A). As the sedimentology of this limestone suggests strong tidal energy, which implies a connection to the open sea, the $\delta^{13}C$ values are interpreted to reflect brackish estuarine conditions[1].

## TECTONOSTRATIGRAPHIC AND PALEOENVIRONMENTAL SETTINGS

### *The UORS Interval*

Although incomplete and not formally measured, the UORS in Bute and Great Cumbrae has a minimum thickness of several hundred metres of sandy to gravelly braidplain deposits, and paleocurrent measurements suggest that both successions were part of the same basin with a source area to the southwest, presumably from a NW-SE trending fault scarp, such as the Sound of Bute Fault (Fig. 13A). This NW-SE trending structure would have acted as a transpressive dextral fault in response to a ~355°-175° shortening direction (*sensu* Coward 1993) (Figs. 13A and 14A).

### *The Kinnesswood Formation Doughend Sandstone Member Interval*

Unconformably overlying the UORS, the Doughend Sandstone Member of the Kinnesswood Formation is of similar thickness at the Great Cumbrae and Bute localities (Fig. 15). It displays similarly oriented paleocurrents to those of the UORS (Fig. 1), suggesting that no significant changes in the tectonostratigraphic setting occurred between the two intervals (Figs. 13A-B and 14A). However, minor readjustment of the fault system may have occurred to account for mild tilting and erosion of the UORS prior to the onset of Kinnesswood Formation deposition, which

[1] Note: because of a nearly complete overlap in $\delta^{18}O$ values for Upper Devonian to Lower Carboniferous calcretes and marine carbonates (Fig. 12), evidence for freshwater and seawater mixing can only be provided by $\delta^{13}C$ values for rocks of that age.

may also account for a slight counter-clockwise rotation of paleocurrent vectors from the UORS to the Inverclyde Group in Great Cumbrae (Fig. 1). During this transition, the sandy to gravelly braidplain setting seemingly evolved towards lower sedimentation rates and dryer conditions that favoured the development of calcretes in the Kinnesswood Formation, which are absent from the underlying UORS at those localities.

### *The Kinnesswood Formation Foul Port Member Interval*

Significant changes occurred in the tectonic setting following deposition of the Doughend Sandstone Member, which resulted in contrasting subsequent stratigraphic successions between the two localities (Fig. 15). It is interpreted that the onset of Foul Port Member deposition corresponds to the development of the Kerrycroy Fault of Caldwell and Young (2013b), which is located between Bute and Great Cumbrae based on geophysical data from McLean and Deegan (1978). This fault delineates the western end of the Cumbraes Trough of Young and Caldwell (2012) (Fig. 1), and as it is nearly parallel to the inferred ~355$^{o}$-175$^{o}$ shortening direction (Coward 1993), it would have acted as a normal growth fault (Figs. 13C and 14B). Base-level rise in response to fault subsidence is interpreted to have caused a transition from sandy-to-gravelly braidplain deposits of the Doughend Sandstone Member to fine grained playa and tidal flat deposits of the Foul Port Member, in which pedogenic oxidation, mottling and calcrete formation provide evidence of prolonged periods of sub-aerial exposure.

**Intermittent marine incursions in a restricted, evaporitic setting.**—Although the thin peritidal carbonate interval near the base of the Foul Port Member is the only clear evidence of a marine incursion in the Cumbraes Trough, the large and ubiquitous dewatering structures in the lower part of that member may provide indirect evidence. The size of these closely stacked structures suggests significant volume loss. However, the fact that they occurred very close to the surface, as indicated by their occasional truncation by sheetflood sandstone, does not suggest that dewatering occurred through compaction. Hence, it is here interpreted that these dewatering structures are associated with the intermittent deposition and dissolution of evaporitic deposits in the lower part of the Foul Port Member. Block subsidence along the Kerrycroy Fault would have accommodated short-lived marine water incursions in the restricted setting of the Cumbraes Trough (Figs. 13C and 14B), followed by extended periods of evaporation, sub-aerial exposure and pedogenesis in a tropical arid, supratidal environment. Intermittent influxes of seawater are also interpreted to be responsible for abnormally high $\delta^{13}C$ values in calcretes of the lower Foul Port Member, which exceed those of the peritidal Doughend Limestone (Fig. 12).

Hence, the lower Foul Port Member is interpreted to have periodically transitioned from marginal marine to continental conditions. Each episode of marine influx or continental freshwater flooding may have caused the dissolution of evaporites and advection of the resulting solutions, preventing long-term evaporite preservation. If such an episode was preceded by a prolonged period of arid pedogenesis, calcrete development in muddy intervals overlying the evaporites would have reduced permeability. In such a scenario, the brines would have been forced to escape in discrete areas, thus creating pseudo-syncline-tepee assemblages (Fig. 5) and, when a greater volume of dissolved evaporites was involved, isolated blocks separated by large water escape pathways (Fig. 6).

It is noteworthy that wavy calcite structures (Fig. 6) only occur in intervals with an apparent absence of calcretic material or other pedogenic features. These intervals may have remained permanently underwater, but alternating between open estuarine and restricted evaporitic conditions, with each marine influx causing the dissolution of previously deposited evaporites. The calcium in these structures possibly originated in part from dissolved sulphates.

**The massive HRPCHs in Bute and Arran**—As noted earlier, thick and massive HRPCHs are quite rare in the geological record, and for this reason, their occurrences in ancient sedimentary basins have been used as stratigraphic markers (e.g., Jutras et al. 1999, 2001, 2007a, b, 2011; Jutras and Prichonnet 2002, 2004, 2005). On the Isle of Arran, near Bute, HRPCH remnants are all found at the same stratigraphic level, disconformably below the Kinnesswood Formation at three localities that otherwise bear contrasting Upper Devonian to Lower Carboniferous successions (Jutras et al. 2011; Young and Caldwell 2012).

The Kinnesswood Formation is much thinner in Arran than in Great Cumbrae. It is charecterized by alternations of dark red mudstone, sandstone and conglomerate with well-developed calcretes and no dewatering structures (Young and Caldwell, 2012) which best correlate with the upper part of the Foul Port Member. Hence, the HRPCH that underlies the Kinnesswood Formation in Arran is possibly contemporaneous with the lower part of the Foul Port Member, in which abundant and exceedingly large dewatering structures suggest a significant loss of volume associated with the dissolution of evaporites.

During deposition of the lower Foul Port Member, episodic marine incursions followed by restriction and evaporation are interpreted to have led to the development of higher pH groundwater in and around the Cumbraes Trough. In contrast, the South Bute Shoulder would have received lower pH fresh groundwater conveyed from subdued highlands to the southwest, which would have mixed with the higher pH groundwater surrounding the evaporitic basin to generate an environment conducive for HRPCHs formation (based on the model of Arakel and McConchie 1982) (Figs. 13D and 14B).

It is noteworthy that the HRPCH at Dunagoil bears unusually high $\delta^{13}C$ values that are not matched by high $\delta^{18}O$ values (Fig. 12) This places it slightly outside the range of other known HRPCHs from the Upper Devonian to Lower Carboniferous successions of Scotland and eastern CanadaJutras et al., 2007b, 2011; Jutras, 2017). The relatively low $\delta^{18}O$ values suggest a climate that was no more arid than that prevailing during the formation of other HRPCH occurrences from that interval, and the high $\delta^{13}C$ values are interpreted to be a consequence of seawater infiltration into the phreatic zone at Dunagoil due to its proximity to the intermittently marine Cumbraes Trough (Fig. 14B). As a result, the HRPCH at Dunagoil bears a $\delta^{13}C$ signature that is similar to what would be expected of brackish water carbonate, such as the peritidal Doughend Limestone marker and estuarine limestone of the Lower Carboniferous Albright Brook beds (Middle Windsor Group) of eastern Canada (Jutras et al. 2007b) (Fig. 12A).

Although Young and Caldwell (2011a) correlated the conglomeratic sandstone that underlies and probably hosts the HRPCH in Bute with the Ballagan Formation based on its dominantly gray colour, it is more likely that these coarse beds were deposited in continuation with the Doughend Sandstone Member succession, but found themselves permanently below the water table shortly after deposition, which would have prevented thorough oxidation. In support of this, the HRPCH occurs at approximately the same stratigraphic level as the Doughend Limestone of Great Cumbrae (Fig. 15), which would have corresponded to a time of regional rise in base-level. Hence, the HRPCH in Bute is here considered to be contemporaneous to the occurrences below the Kinnesswood Formation in Arran, with the latter succession being possibly contemporaneous to the upper part of the Foul Port Member in Great Cumbrae.

In summary, the absence of Foul Port Member strata at Dunagoil Bay can be explained by its location outside of the Cumbraes Trough at the time of deposition, and formation of the HRPCH that tops the Doughend Sandstone Member in Bute resulted from proximity of the succession on this shoulder to the intermittently evaporitic basin in which the lower part of the Foul Port Member is interpreted to have been deposited. Such correlation provides more precise constraints on the timing of HRPCH formation in both Arran and Bute, shortly after deposition of the Doughend Sandstone Member, and synchronous with deposition of the lower part of the Foul Port Member. Because of the stratigraphic significance of the HRPCHs in Arran and Bute, it is here proposed to formalize these penecontemporaneous occurrences as the Dunagoil Calcrete, a lithodemic sub-unit of the Kinnesswood Formation.

## CONCLUSIONS

Based on facies distribution, provenance and paleocurrent data, the Upper Devonian to Lower Carboniferous Inverclyde Group in Bute and Great Cumbrae was sourced from fault-bounded highlands occupied by Dalradian basement rocks and some possible remnants of the Lower Old Red Sandstone to the southwest (Figs. 13 and 14). Correlations between the two isles suggest that HRPCH formation on the Bute Shoulder occurred following the development of the Cumbraes Trough along an intra-basinal growth fault (the Kerrycroy Fault) at an interval corresponding to the lower half of the Famennian Kinnesswood Formation Foul Port Member. Based on the presence of a peritidal carbonate bed in the Cumbraes Trough at the base of this interval, the rest of which is pervasively disturbed by large dewatering structures interpreted to be the result of dissolved evaporite withdrawal, the latter trough accommodated intermittent seawater incursions that were followed by restriction and evaporitic conditions. This intermittent evaporitic basin is inferred to be responsible for the formation of a HRPCH on the adjacent Bute Shoulder, where relatively low pH fresh groundwater issued from subdued highlands to the southwest would have mixed with higher pH groundwater surrounding the evaporitic basin, thus creating an increase in silicate solubility paired with a decrease in calcium carbonate solubility, which resulted in the thorough replacement of silicates by phreatic calcrete.

Stratigraphic correlation of the HRPCH interval on the Bute Shoulder with beds that show evidence for the former presence of evaporites in the Cumbraes Trough corroborates previous studies in which a close connection to an evaporitic basin is considered to be necessary for the formation of thick and massive occurrences of such calcretes (e.g., Arakel and McConchie 1982; Jutras et al. 2007b). It also strengthens the argument that such calcretes can be used as indicators for the former presence in their vicinity of evaporites (e.g., Jutras et al. 2011; Jutras 2017), which are far less likely to be preserved in the geological record. Hence, it is inferred that the HRPCH occurrences on the northwestern and northeastern shores of Arran, which are at the same approximate stratigraphic level (Jutras et al. 2011), developed in a similar setting as the HRCPH in Bute, possibly in the vicinity of other troughs that hosted evaporitic bodies. However, based on its higher $\delta^{13}C$ values, which resemble those of brackish water carbonates, the HRPCH in Bute may have developed closer to an intermittently marine basin, and its aquifer may have received occasional influxes of seawater during its formation.

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## FIGURE CAPTIONS

FIG. 1.—Simplified geology of the Firth of Clyde area of southwest Scotland (modified from Young and Caldwell 2012), with paleocurrent data from trough channel orientations in the UORS (orange rose diagrams) and the Inverclyde Group (yellow rose diagrams). GC: Great Cumbrae; LC: Little Cumbrae.

FIG. 2.—Upper Devonian to Lower Carboniferous stratigraphy of the Firth of Clyde area based on Young and Caldwell (2011b) and Marshall et al. (2019). Time scale after Herbig et al. (2018).

FIG. 3.—Measured sections in Great Cumbrae, with sampled intervals.

FIG. 4.—**A)** Brownish-red mudstone with calcrete nodules transitioning upward into gray marl (at the base of the pen) and intrasparudite (at the top of the pen) of the Doughend Limestone marker near the base of the lower Foul Port Member along the southwest coast of Great Cumbrae. **B)** Interference ripple marks enhanced by dewatering structures. Pen for scale.

FIG. 5.—–Large “pseudo-synclines” (red dotted lines) separated by “tepee” structures in brownish-red mudrock of the lower part of the Foul Port Member in its type-area of Foul Port, Great Cumbrae.

FIG. 6.—–Isolated blocks of calcretized mudstone (1) separated by large dewatering pathways in the lower part of the Foul Port Member in its type-area of Foul Port, Great Cumbrae.

FIG. 7.—–Muddy intervals with wavy calcite truncated by sandy sheetflood deposits in the lower part of the Foul Port Member in its type-area of Foul Port, Great Cumbrae.

FIG. 8.—–Pseudo-anticline formed by invasive phreatic calcrete lenses (1-3) developed between muddy aquicludes in the Ballagan Formation near Foul Port, Great Cumbrae.

FIG. 9.—–Measured section at Dunagoil, Isle of Bute, with sampled intervals (legend in Figure 3). HRPCH: host-replacing phreatic calcrete hardpan.

FIG. 10.—–**A)** Low-angle unconformity between the Upper Old Red Sandstone (UORS) and the Doughend Sandstone Member of the Kinnesswood Formation, with **B)** syn-Kinnesswood calcrete nodules extending below the unconformity. **C)** Satellite image (from Google Earth) of the Famennian to Lower Carboniferous succession at Dunagoil Bay, southwest Bute.

Fig. 11.—–Partly preserved detrital quartz grain floating in the massive, micritic matrix that forms the bulk of the host-replacing phreatic calcrete hardpan at Dunagoil Bay, southwest Bute. Such remnants of the host sediment form less than 0.5% of the calcrete volume.

FIG. 12.—–$\delta^{13}C$ VPDB vs $\delta^{18}O$ VPDB values in calcretes and carbonate rocks of the Inverclyde Group on the isles of Bute (filled symbols) and Great Cumbrae (open symbols). The data are plotted in relationship with the ranges of Upper Devonian to Lower Carboniferous marine

carbonates, worldwide, and calcretes of all types from the same interval in Scotland and eastern Canada based on previous studies, with the addition in **A)** of the more specific range of host-replacing phreatic calcrete hardpans (HRPCHs). Also indicated is the range of HRPCHs in Quaternary successions of Western and Central Australia based on Jacobson et al. (1988). **B)** presents the data in its stratigraphic context. Notes: samples with abnormally low $\delta^{18}O$ values were excluded from the ranges; 1: estuarine Visean carbonate from eastern Canada (Jutras et al. 2007b); 2: Mora et al. (1991) provided some lower $\delta^{13}C$ values (-8.69 ±1.56; N = 12) in calcretes from the Upper Devonian Catskills Formation of Pennsylvania, but did not provide $\delta^{18}O$ values. VPDB: Vienna Pee Dee Belemnite.

FIG. 13.—Famennian paleogeographic reconstructions of the south Bute (1) and Great Cumbrae (2) areas during deposition of sandy to gravelly braidplain deposits of **A)** the Upper Old Red Sandstone and **B)** the Kinnesswood Formation Doughend Sandstone Member, as well as during deposition of **C)** the Doughend Limestone marker and **D)** tidal flat deposits of the lower part of the Kinnesswood Formation Foul Port Member, with syn-depositional formation of a host-replacing phreatic calcrete hardpan on the South Bute Shoulder.

FIG. 14.—Reconstructed cross-section (not to scale) showing **A)** basin development along a transpressional dextral fault (Sound of Bute Fault?) during deposition of the Famennian Upper Old Red Sandstone and Kinnesswood Formation Doughend Sandstone Member, and **B)** subsequent development of the Cumbraes Trough along a growth fault (the Kerrycroy Fault) during deposition of the Kinnesswood Formation Foul Port Member playa and tidal flat deposits. The latter subbasin accommodated intermittent sea incursions followed by restriction and evaporation, which allowed the development of a host-replacing phreatic calcrete hardpan in the previously deposited Doughend Member near the trough, where mixing of low pH fresh

groundwater and high pH evaporitic groundwater would have occurred. The inferred lateral transition from phreatic calcrete to gypcrete is based on the model of Arakel and McConchie (1982).

FIG. 15.—Summary sections of the Famennian to Lower Carboniferous successions on the South Bute Shoulder (Dunagoil section) and Cumbraes Trough (southwest coast and Foul Port sections). Legend in Figure 3.

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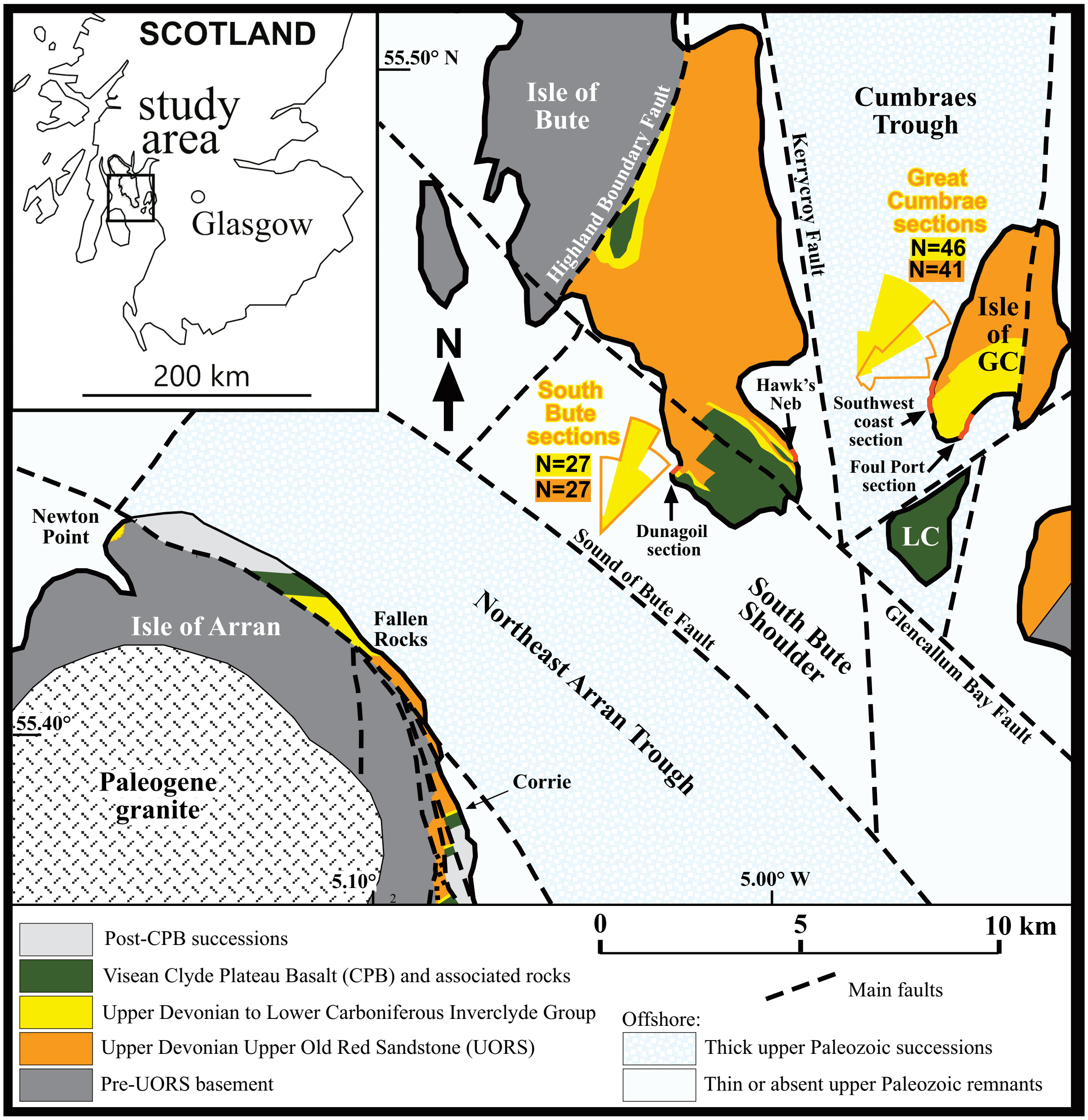
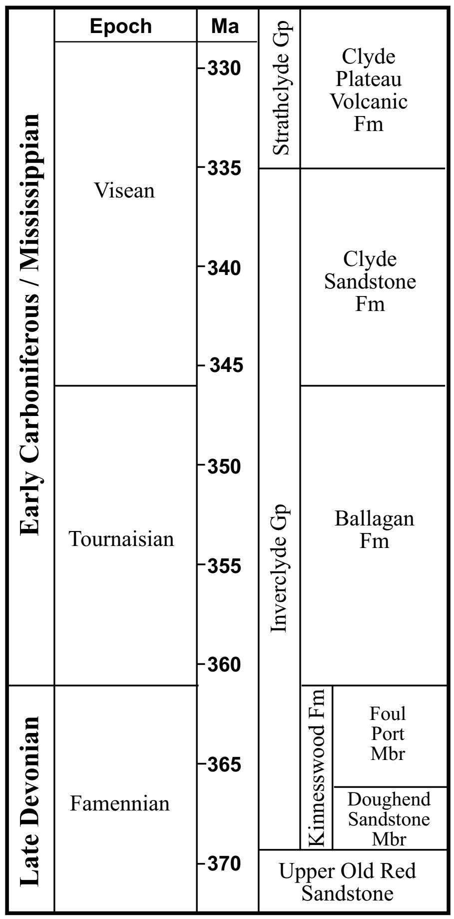
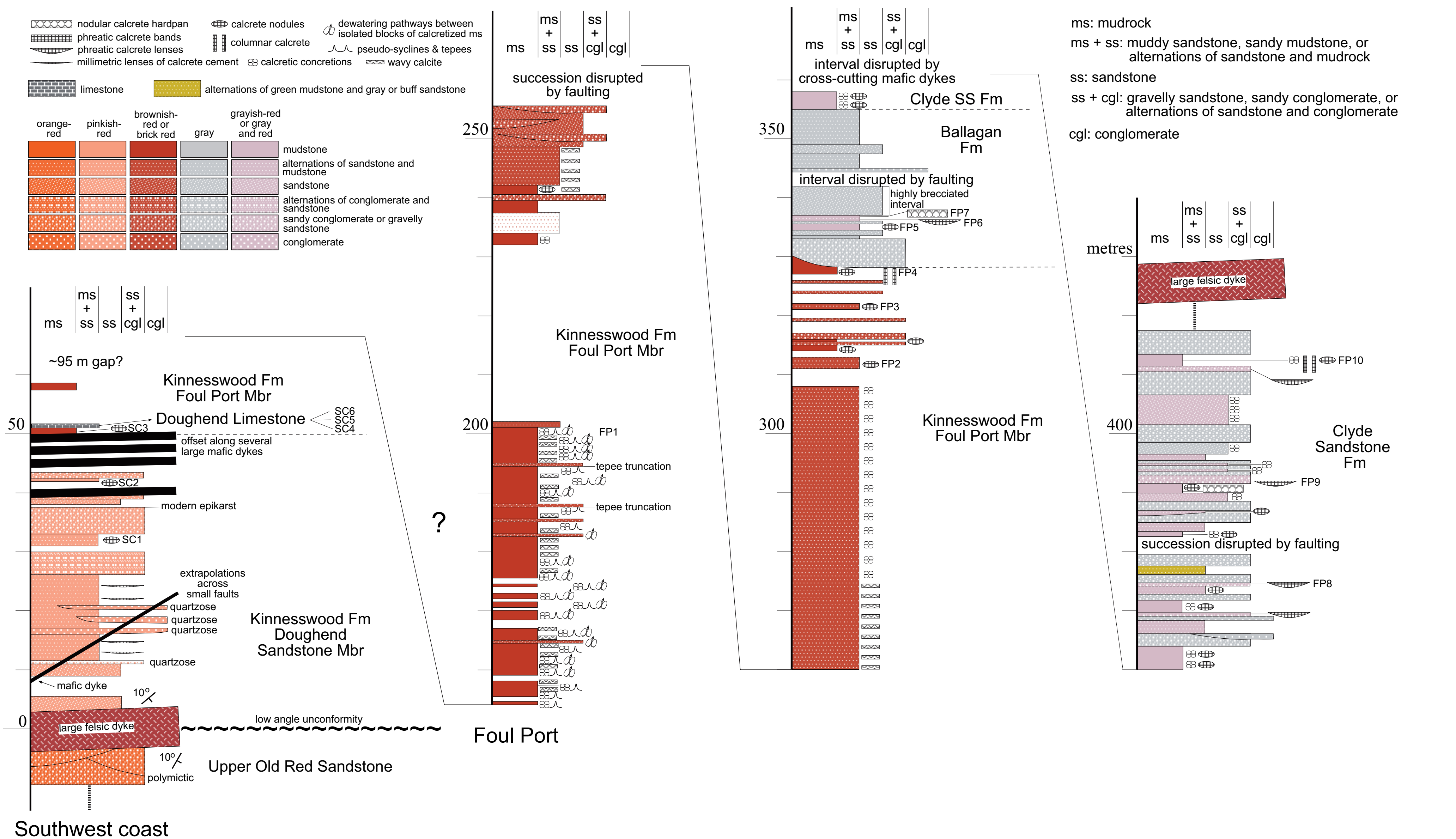
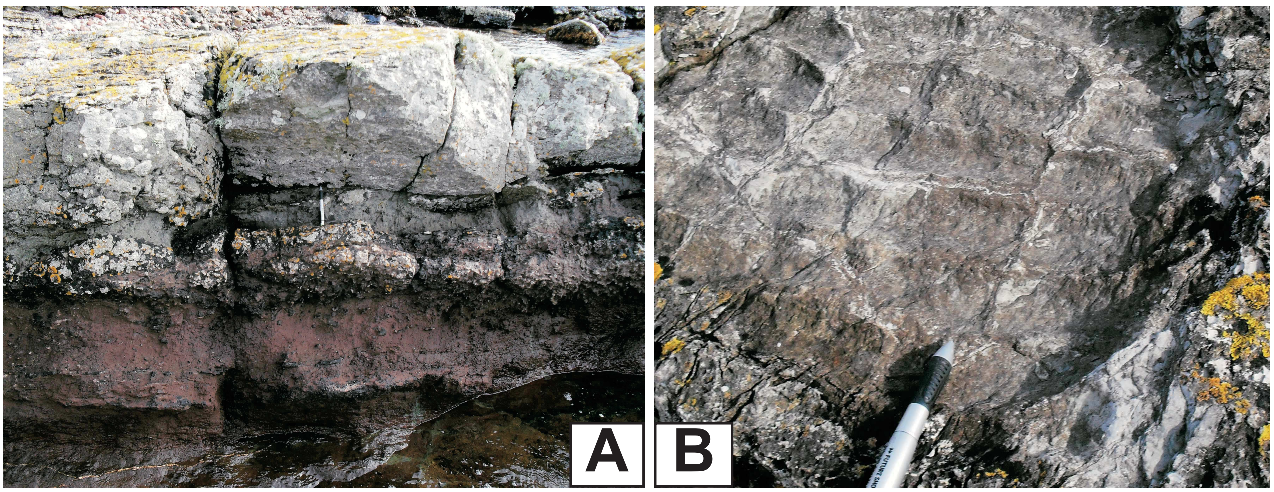
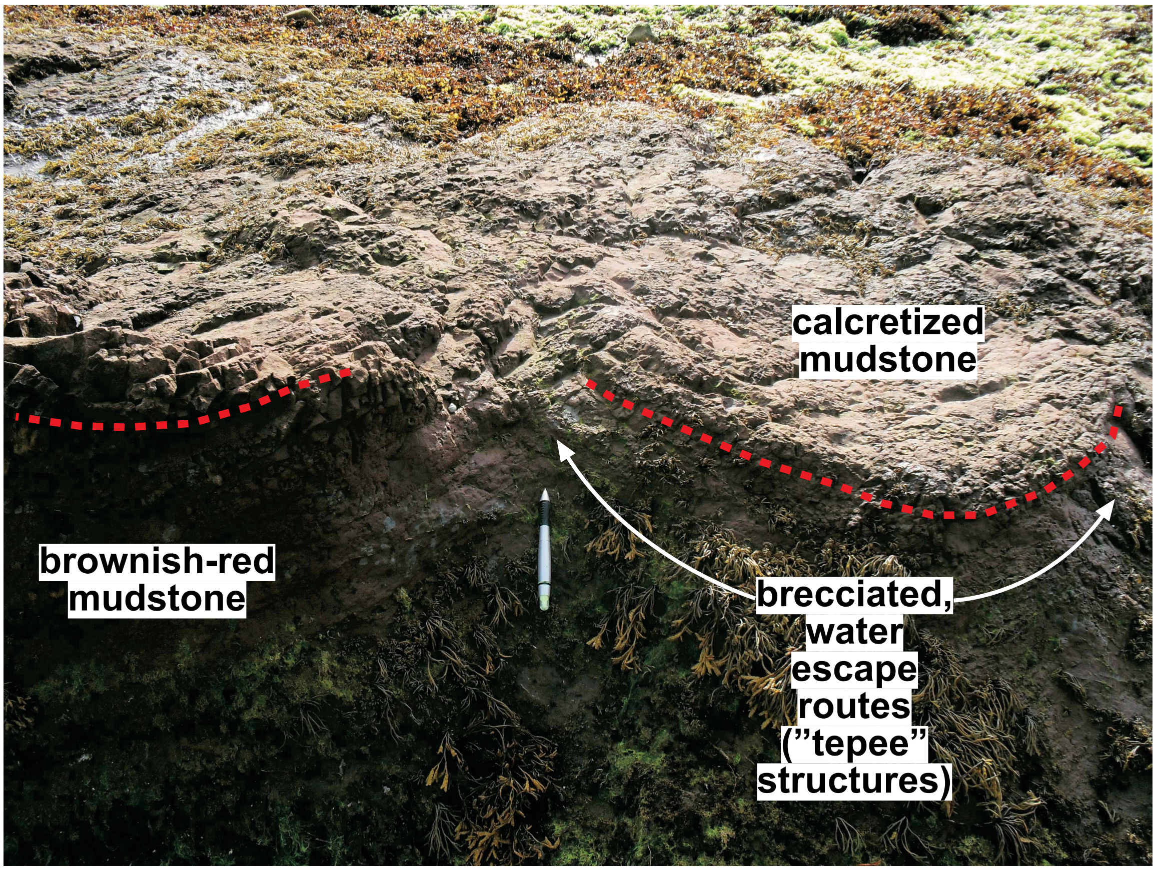
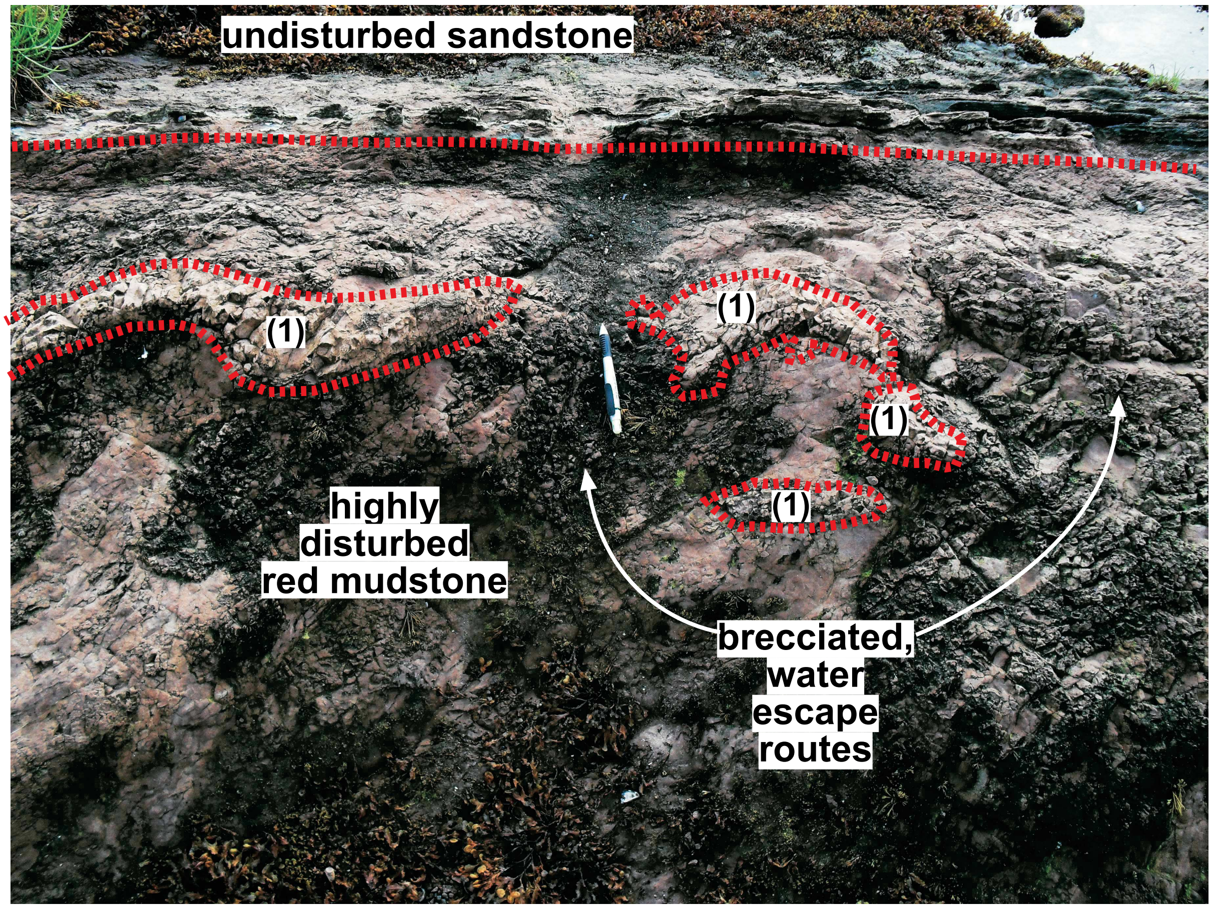

SCOTLAND
study area
Glasgow
200 km
55.50° N
Isle of Bute
Highland Boundary Fault
Kerrycroy Fault
Cumbraes Trough
Great Cumbrae sections
N=46
N=41
Isle of GC
N
South Bute sections
N=27
N=27
Hawk's Neb
Southwest coast section
Foul Port section
Dunagoil section
LC
Newton Point
Isle of Arran
Fallen Rocks
Sound of Bute Fault
Northeast Arran Trough
South Bute Shoulder
Glencallum Bay Fault
55.40°
Paleogene granite
Corrie
5.10°
5.00° W
0
5
10 km
Main faults
Post-CPB successions
Visean Clyde Plateau Basalt (CPB) and associated rocks
Upper Devonian to Lower Carboniferous Inverclyde Group
Upper Devonian Upper Old Red Sandstone (UORS)
Pre-UORS basement
Offshore:
Thick upper Paleozoic successions
Thin or absent upper Paleozoic remnants

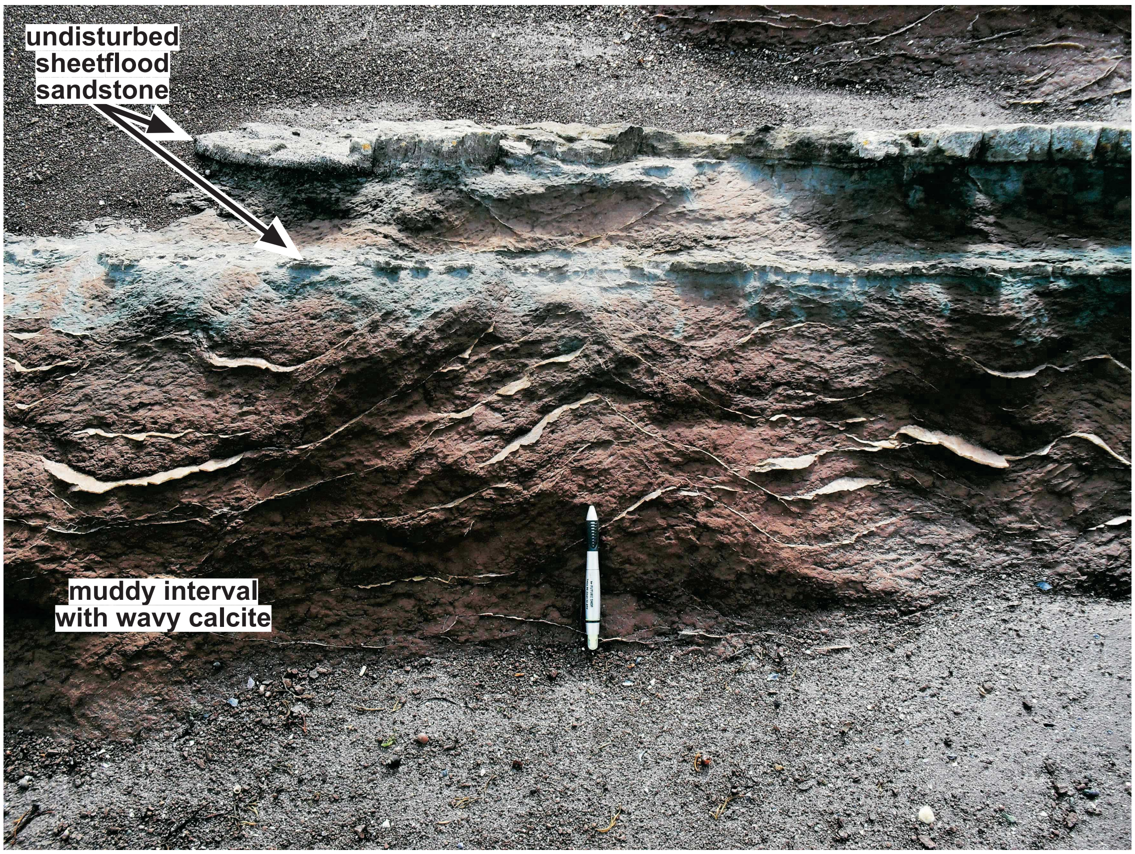

Early Carboniferous / Mississippian
Late Devonian
Epoch
Ma
Visean
Tournaisian
Famennian
330
335
340
345
350
355
360
365
370
Strathclyde Gp
Inverclyde Gp
Clyde Plateau Volcanic Fm
Clyde Sandstone Fm
Ballagan Fm
Kinnesswood Fm
Foul Port Mbr
Doughend Sandstone Mbr
Upper Old Red Sandstone

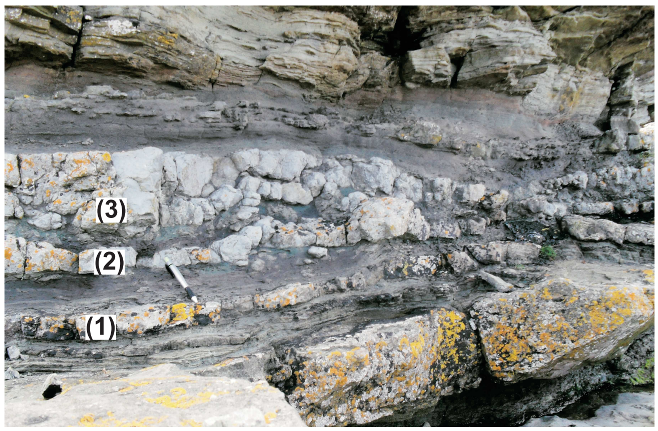

nodular calcrete hardpan
phreatic calcrete bands
phreatic calcrete lenses
millimetric lenses of calcrete cement
calcrete nodules
columnar calcrete
calcretic concretions
dewatering pathways between isolated blocks of calcretized ms
pseudo-synclines & tepees
wavy calcite
limestone
alternations of green mudstone and gray or buff sandstone
orange-red
pinkish-red
brownish-red or brick red
gray
grayish-red or gray and red
mudstone
alternations of sandstone and mudstone
sandstone
alternations of conglomerate and sandstone
sandy conglomerate or gravelly sandstone
conglomerate
ms: mudrock
ms + ss: muddy sandstone, sandy mudstone, or alternations of sandstone and mudrock
ss: sandstone
ss + cgl: gravelly sandstone, sandy conglomerate, or alternations of sandstone and conglomerate
cgl: conglomerate
ms
ms + ss
ss
ss + cgl
cgl
~95 m gap?
Kinnesswood Fm Foul Port Mbr
Doughend Limestone
SC6
SC5
SC4
SC3
offset along several large mafic dykes
SC2
modern epikarst
SC1
extrapolations across small faults
quartzose
Kinnesswood Fm Doughend Sandstone Mbr
mafic dyke
10°
large felsic dyke
low angle unconformity
polymictic
Upper Old Red Sandstone
Southwest coast
0
50
?
succession disrupted by faulting
250
200
FP1
tepee truncation
Kinnesswood Fm Foul Port Mbr
Foul Port
interval disrupted by cross-cutting mafic dykes
Clyde SS Fm
350
Ballagan Fm
interval disrupted by faulting
highly brecciated interval
FP7
FP6
FP5
FP4
FP3
FP2
300
Kinnesswood Fm Foul Port Mbr
metres
large felsic dyke
FP10
400
Clyde Sandstone Fm
FP9
succession disrupted by faulting
FP8

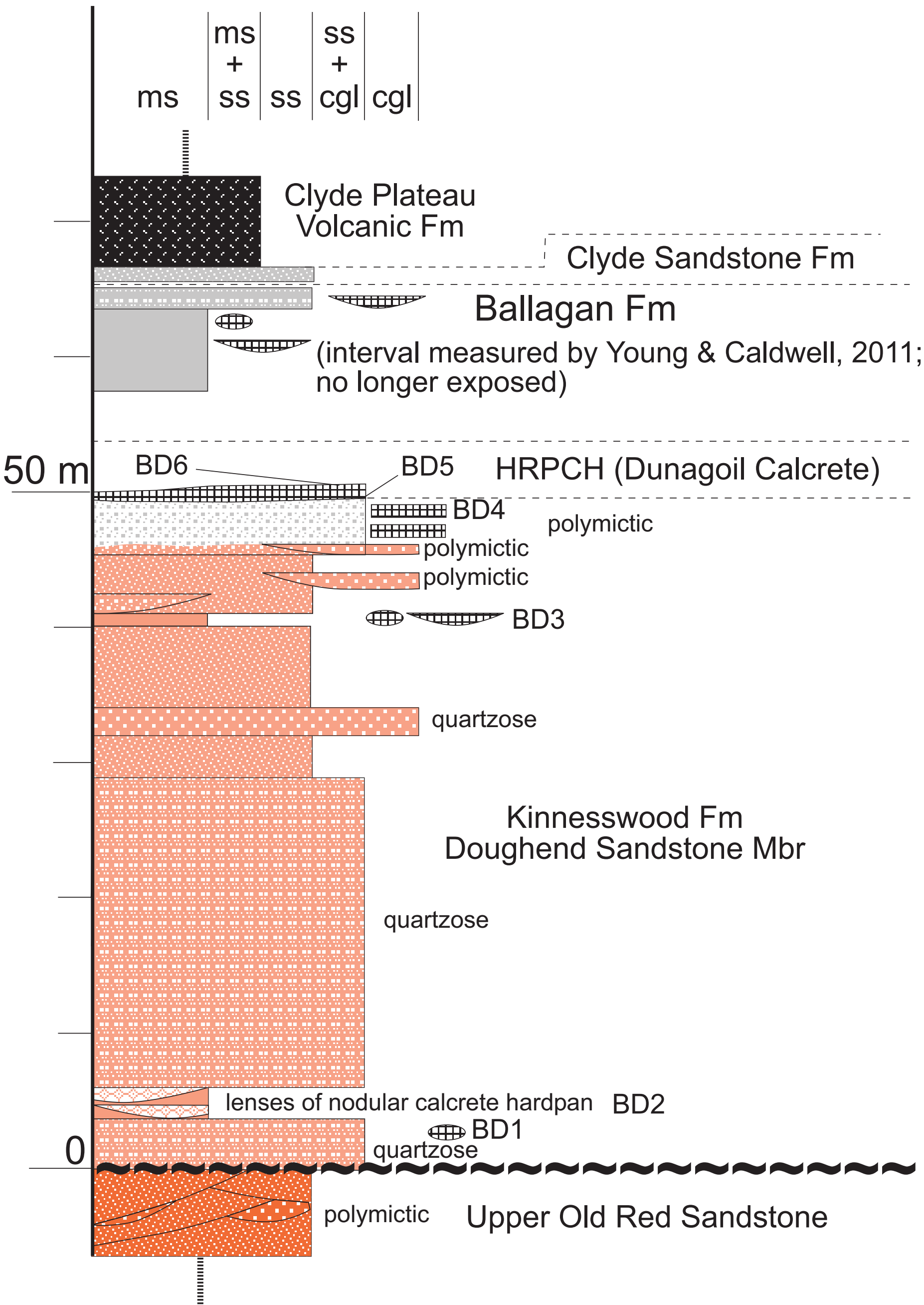

A
B

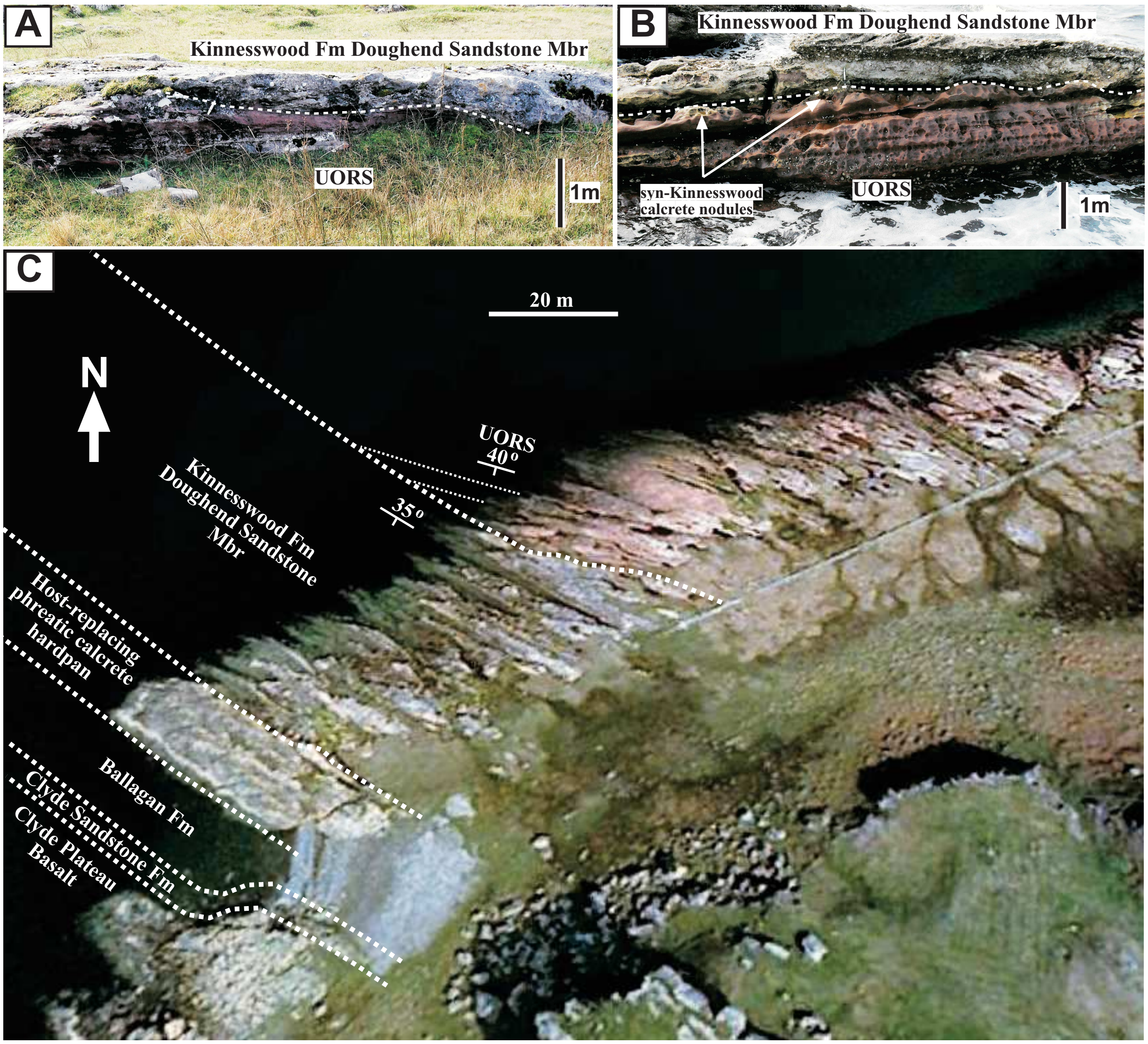

calcretized
mudstone
brownish-red
mudstone
brecciated,
water
escape
routes
("tepee"
structures)

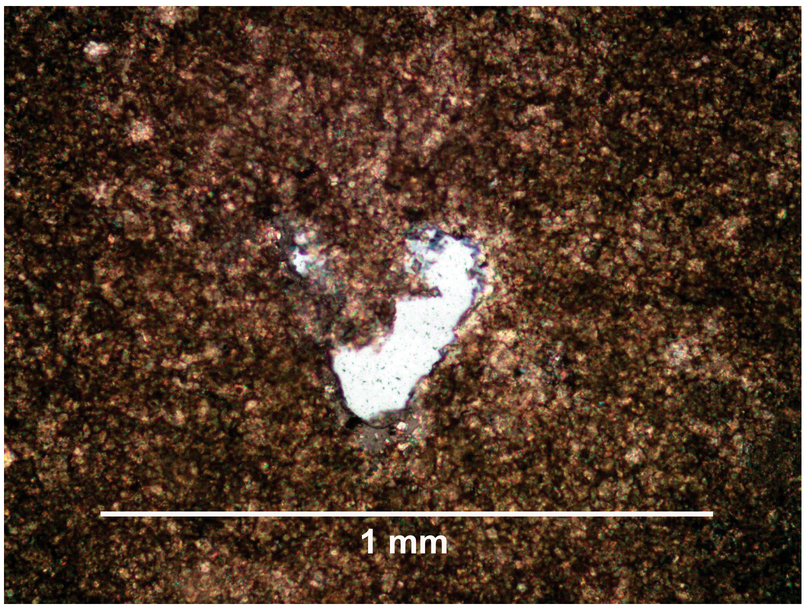

undisturbed sandstone
(1)
(1)
(1)
(1)
highly
disturbed
red mudstone
brecciated,
water
escape
routes


undisturbed
sheetflood
sandstone
muddy interval
with wavy calcite

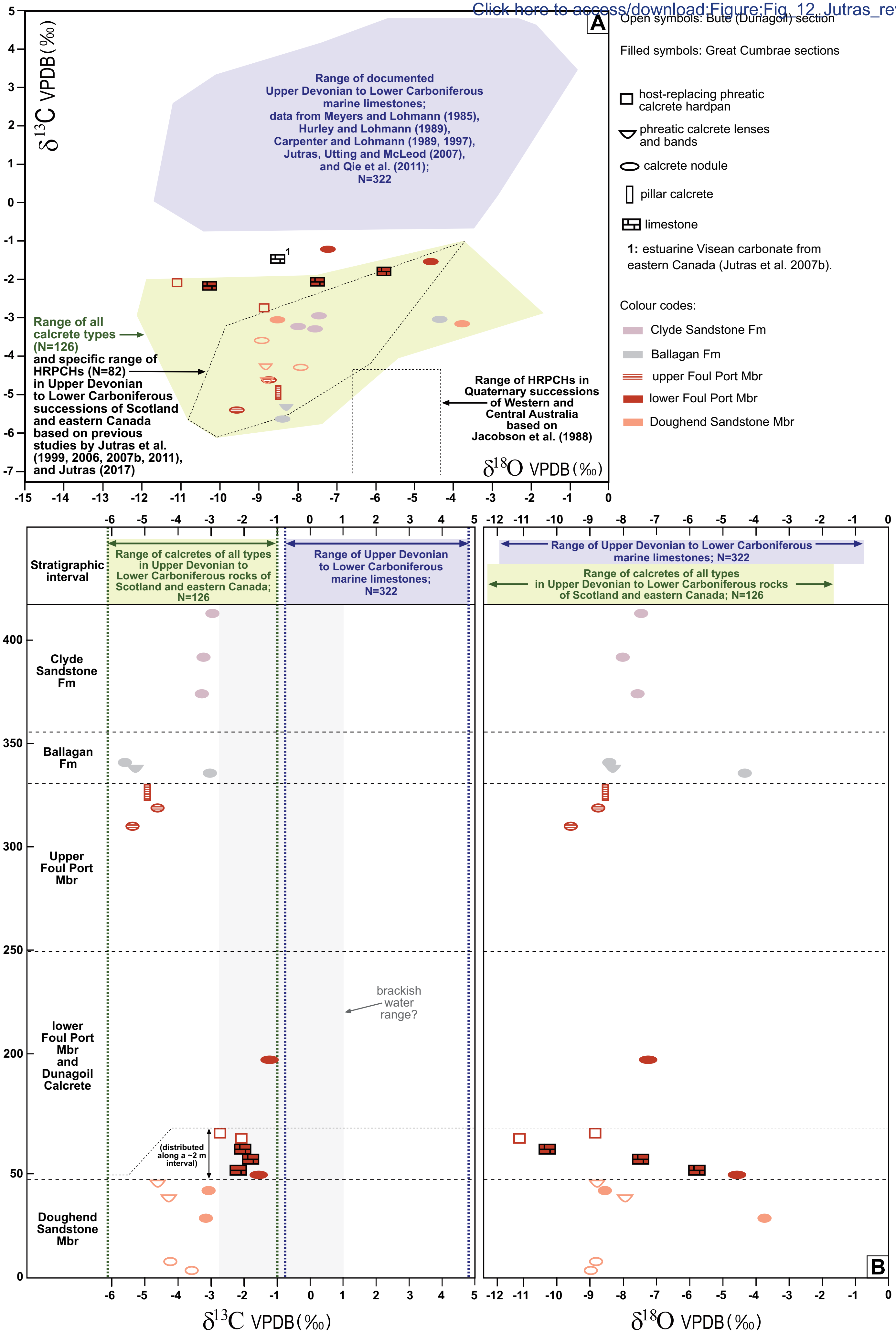

(3)
(2)
(1)

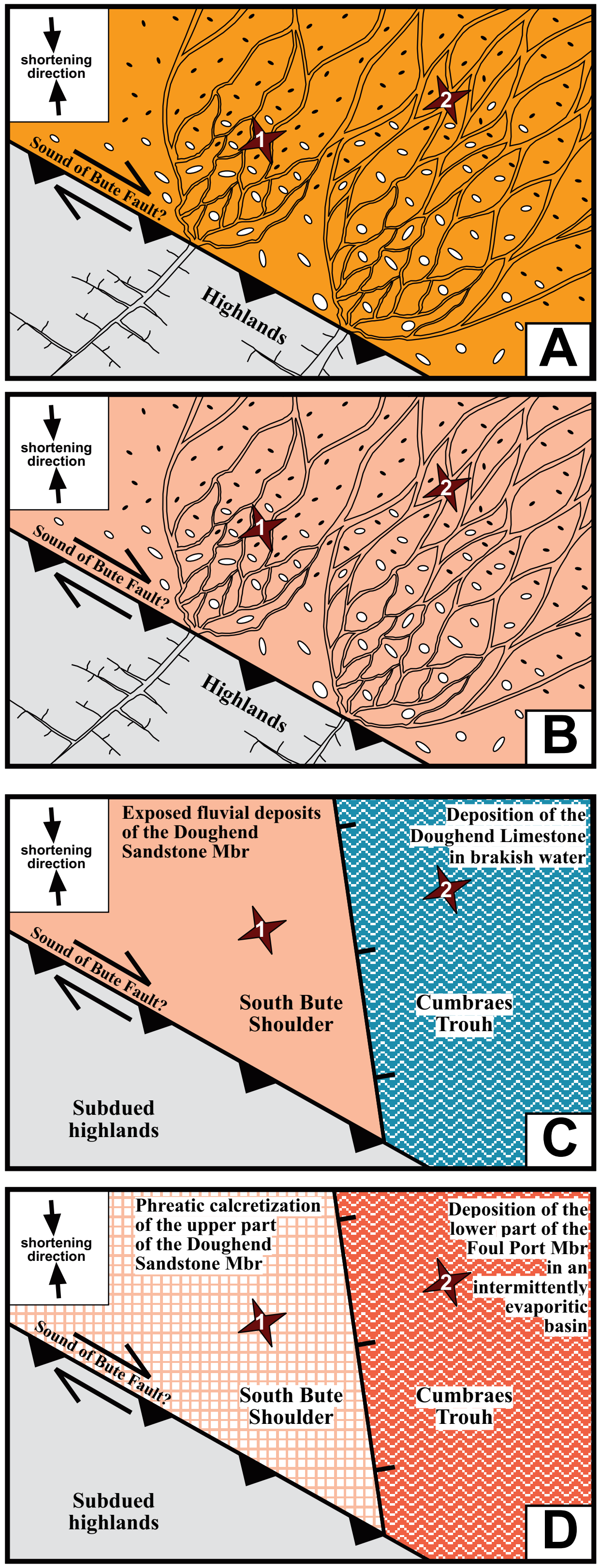

ms
ms + ss
ss
ss + cgl
cgl
Clyde Plateau Volcanic Fm
Clyde Sandstone Fm
Ballagan Fm
(interval measured by Young & Caldwell, 2011; no longer exposed)
50 m
BD6
BD5
HRPCH (Dunagoil Calcrete)
BD4
polymictic
polymictic
polymictic
BD3
quartzose
Kinnesswood Fm
Doughend Sandstone Mbr
quartzose
lenses of nodular calcrete hardpan
BD2
BD1
quartzose
0
polymictic
Upper Old Red Sandstone

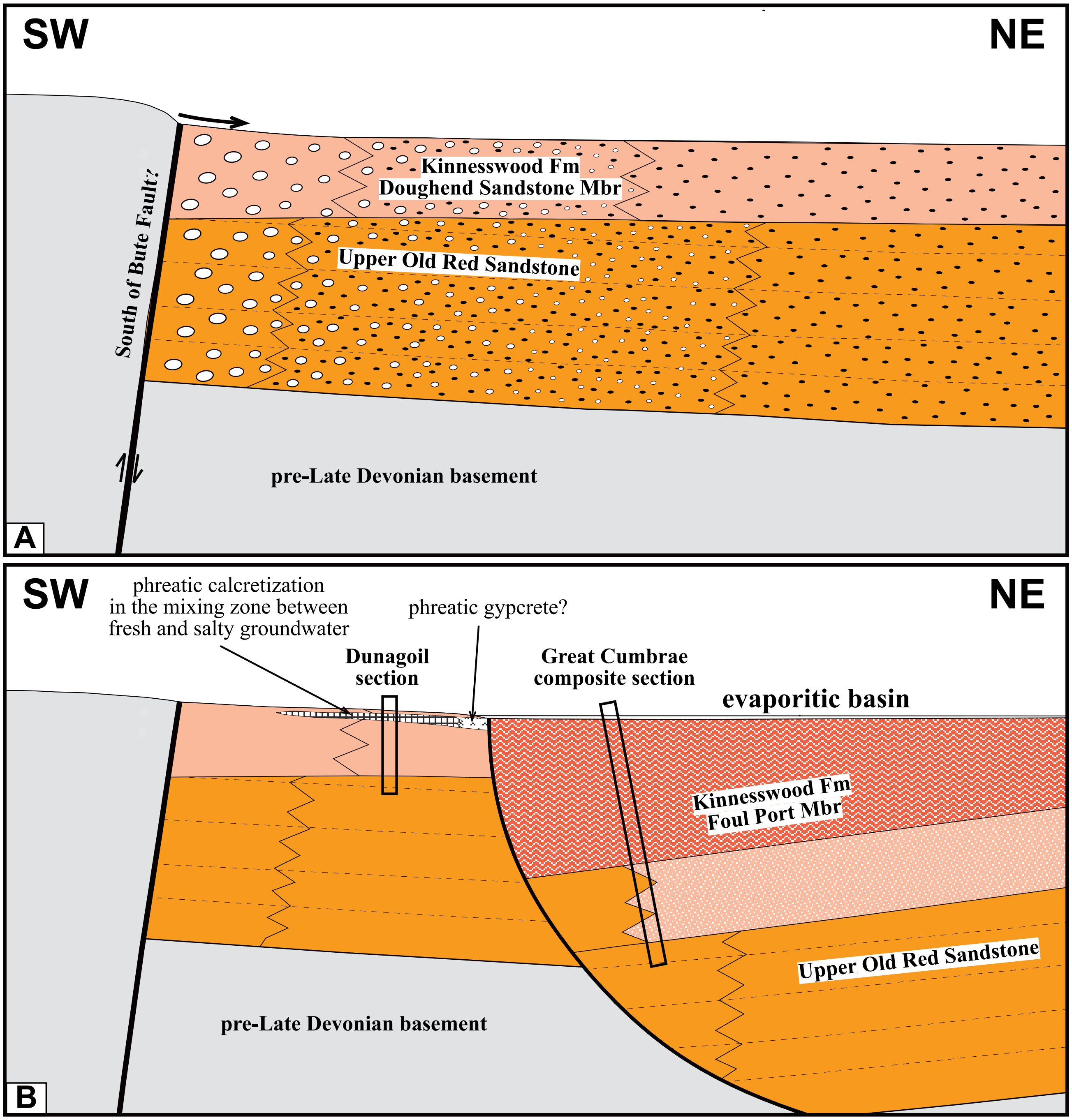

A
Kinnesswood Fm Doughend Sandstone Mbr
UORS
1m
B
Kinnesswood Fm Doughend Sandstone Mbr
syn-Kinnesswood calcrete nodules
UORS
1m
C
20 m
N
UORS
40°
35°
Kinnesswood Fm Doughend Sandstone Mbr
Host-replacing phreatic calcrete hardpan
Ballagan Fm
Clyde Sandstone Fm
Clyde Plateau Basalt

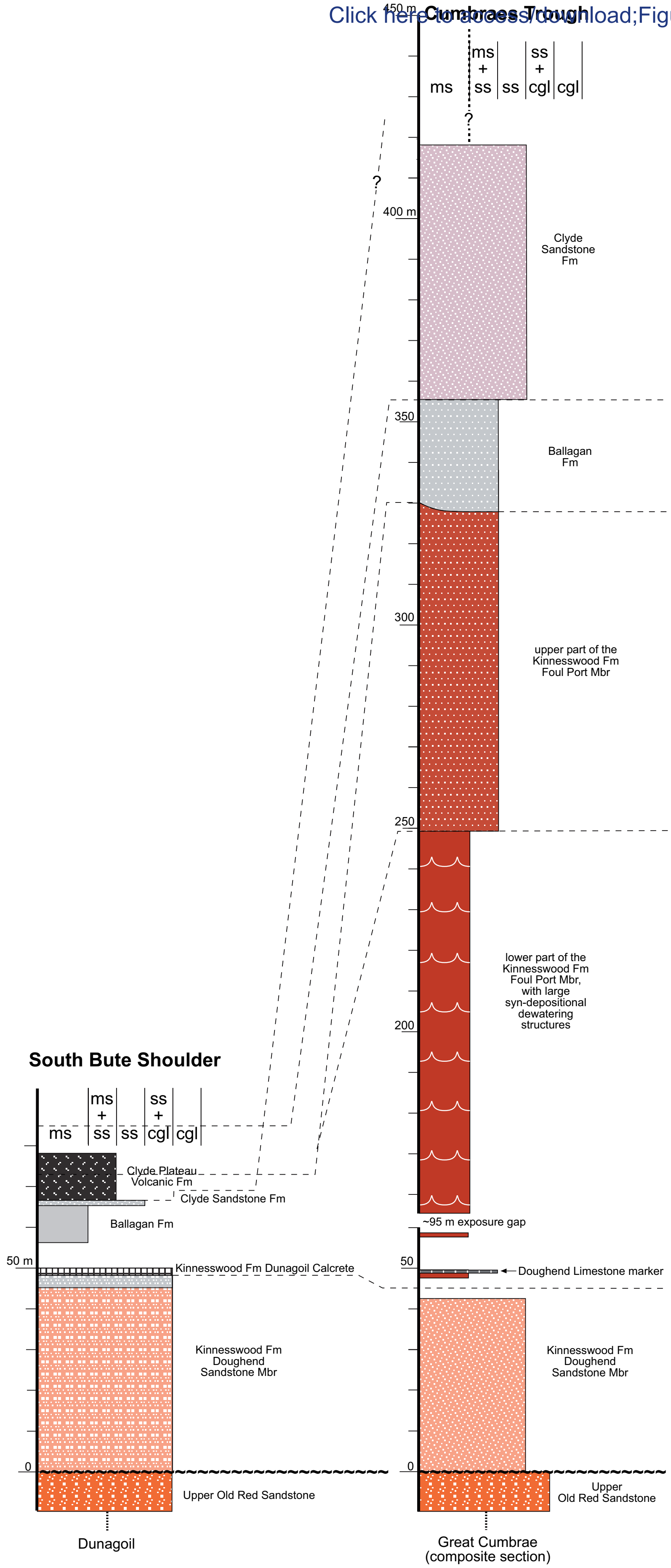

1 mm

Click here to access/download;Figure;Fig_12_Jutras_revised.pdf




shortening direction
Sound of Bute Fault?
Highlands
1
2
A
shortening direction
Sound of Bute Fault?
Highlands
1
2
B
shortening direction
Exposed fluvial deposits of the Doughend Sandstone Mbr
Deposition of the Doughend Limestone in brakish water
Sound of Bute Fault?
South Bute Shoulder
Cumbraes Trouh
Subdued highlands
1
2
C
shortening direction
Phreatic calcretization of the upper part of the Doughend Sandstone Mbr
Deposition of the lower part of the Foul Port Mbr in an intermittently evaporitic basin
Sound of Bute Fault?
South Bute Shoulder
Cumbraes Trouh
Subdued highlands
1
2
D


SW
NE
Kinnesswood Fm
Doughend Sandstone Mbr
Upper Old Red Sandstone
South of Bute Fault?
pre-Late Devonian basement
A
SW
NE
phreatic calcretization
in the mixing zone between
fresh and salty groundwater
phreatic gypcrete?
Dunagoil
section
Great Cumbrae
composite section
evaporitic basin
Kinnesswood Fm
Foul Port Mbr
Upper Old Red Sandstone
pre-Late Devonian basement
B


Cumbraes Trough
450 m
ms
ms + ss
ss
ss + cgl
cgl
?
?
400 m
Clyde Sandstone Fm
350
Ballagan Fm
300
upper part of the Kinnesswood Fm Foul Port Mbr
250
lower part of the Kinnesswood Fm Foul Port Mbr, with large syn-depositional dewatering structures
200
~95 m exposure gap
50
Doughend Limestone marker
Kinnesswood Fm Doughend Sandstone Mbr
0
Upper Old Red Sandstone
Great Cumbrae (composite section)
South Bute Shoulder
ms
ms + ss
ss
ss + cgl
cgl
Clyde Plateau Volcanic Fm
Clyde Sandstone Fm
Ballagan Fm
50 m
Kinnesswood Fm Dunagoil Calcrete
Kinnesswood Fm Doughend Sandstone Mbr
0
Upper Old Red Sandstone
Dunagoil