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# Silcretes: insights into the occurrences and formation of materials sourced for stone tool making

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**Abstract**

Silcretes are clearly observed and abundant as components of paleolandscapes on several continents. Mechanisms for the formation of several varieties of silcrete, with specific relationships to paleolandscapes, are described. Each type of silcrete displays particular morphological features in its profile in the paleo-regolith, and these features provide pointers to its origin via mechanisms of *absolute* or *relative* accumulation of silica in specific environments relating to groundwater or soil-water hydrology. The characters of silcrete varieties that may have triggered the interest of prehistoric peoples to exploit them for manufacturing stone tools, and which control knappability, include granulometry and the specific nature of silica cements. The successions of silica precipitation and recrystallisation events are clearly evident as a complex of micromorphological features that provide clues to the hydrological environment and its geochemistry at the time or times of silicification. Examples are given of the distribution of different silcrete facies, which could have had differing values for exploitation for stone tool production, in modern-day landscapes in France and Australia.

*Highlights*

Silcretes occur widely in paleolandscapes in several continents.  
What are they and how did they form?  
What made silcretes suitable for prehistoric people to form stone tools?

*Keywords*

silicification, silcrete, petrography, mineralogy, landscapes, prehistoric tools.

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## 36 1 Introduction

37 There is a wealth of literature on silcretes in both the geological sciences and the  
38 archaeological sciences. In geology it ranges from initial compilations of knowledge like that  
39 edited by Langford-Smith (1978) to the comprehensive overview of Nash and Ulliyot (2007).  
40 In archaeology in Australia the studies described by Holdaway and Fanning (2014) also  
41 provide a review of current knowledge and a foundation for future research. The science  
42 pertaining to silcretes has actually advanced in parallel in both scientific fields with only  
43 limited cross-reference, except perhaps for the work of Webb and his colleagues (Webb and  
44 Domanski, 2008). In geology the early work focussed on field observations and relationships,  
45 and progressed to petrographic and mineralogical studies, from which hypotheses were  
46 generated about origin and palaeoenvironmental conditions. From our geological perspective,  
47 some studies of stone tools in archaeological science had a ‘primitive’ view of silcrete, such  
48 as that expressed in Mulvaney & Kamminga (1999, p 213), but others developed a  
49 sophisticated understanding of the rock mechanical properties of silcrete and other siliceous  
50 materials (Domanski *et al.*, 1994), and some also used petrographic/micromorphological  
51 fabrics and textures (e.g. Summerfield, 1983) to characterise silcrete artefacts and proffer  
52 ideas about their provenance. But the connection with step-by-step advances in our  
53 understanding of the complex of processes and environmental conditions that have led to the  
54 formation of silcretes has not been maintained and the aim of this paper is to at least partly  
55 address this situation.

56 In the first instance, we should be clear about terminology. There are several  
57 definitions of the term ‘silcrete’ but we prefer something along the lines of that proposed by  
58 Eggleton (2001), namely;

59 *‘Strongly silicified, indurated regolith, generally of low permeability, commonly*  
60 *having a conchoidal fracture with a vitreous lustre. Represents the complete or near-*  
61 *complete silicification of regolith by the transformation of precursor silica or silicates*  
62 *and/or the infilling of available voids, including fractures. On a macroscopic scale,*  
63 *some silcretes are dense and massive, but others may be nodular, columnar, blocky,*  
64 *or cellular with boxwork structure. On a microscopic scale, the fabric, mineralogy*

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65            *and composition of silcretes may reflect those of the parent material, but also indicate*  
66            *the changes experienced by, as well as the general environments of, silicification.*

67            The key component of the definition, from our perspective, is the phrase ‘*strongly*  
68            *indurated, silicified regolith*’. Thus, silica-rich secondary materials including flint, chert,  
69            agate, chalcedony, and precious and common opal are not silcretes, though there may be a  
70            spatial and temporal association with some forms of silcrete. Sandstone, quartzite and some  
71            forms of porcellanite and jasper (as defined, for example, in Gary *et al.*, 1973) that have  
72            formed specifically as a result of secondary silicification of the regolith are silcretes, but those  
73            resulting from diagenetic alteration or metamorphism are not. Red-brown hardpans (Chartres,  
74            1985; Wright, 1983) are not silcretes because they are generally not ‘*strongly indurated*’ by  
75            silica.

76            It is also important to note that the term ‘*silcrete*’ is an English word and concept that  
77            has no equivalent in the geosciences literature in Latin languages where such material is  
78            generally called ‘*sandstone*’, ‘*silicified limestone*’, ‘*silicified claystone*’, and so on. In France,  
79            the word ‘*silcrete*’ was used for the first time by Parron *et al.* (1976). As well, ‘*flint tool*’ is  
80            often used as a generic term in the French literature and could have been formed from  
81            silicified limestone or pedogenic silcrete.

82            The environments of formation and ages of silcretes have been the subject of many  
83            investigations over many years, but definitive answers still elude us. Recent work in  
84            Australia on the formation and age of precious opal in Great Australian Basin landscapes  
85            (Dutkiewicz *et al.*, 2015; Rey, 2013; see also critique by Dickson, 2014 and response by Rey,  
86            2014) did not unravel the genetic and temporal relationship between the widespread formation  
87            of opal in voids and fractures in the regolith (within and outside the margins of the Great  
88            Australian Basin) and the complex suite of silcrete facies in these regions. Some work on  
89            O/H isotope composition and trace element chemistry of opal and the siliceous matrix in  
90            silcretes (Alexandre *et al.*, 2004; Dutkiewicz *et al.*, 2015; Harwood *et al.*, 2013; Webb and  
91            Golding, 1998), may provide a future basis for interpreting conditions of formation, though  
92            not age.

93            In this paper we summarise our current thinking on the characteristics, landscape  
94            associations, origin and age of silcretes. We also attempt to link petrological and

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95 mineralogical features (which are of specific importance to geologists in generating  
96 hypotheses to explain the origin of silcretes) to the potential value of silcretes to prehistoric  
97 peoples for the production of stone tools,. However, we recognise that there are substantial  
98 fields of knowledge in geoarchaeology and lithic technology that rightly lay claim to the  
99 development of an understanding of the associations between prehistoric peoples and the  
100 source materials for stone tools.

101        Numerous siliceous materials used for tool making (flint, chert, quartzite, quartz) are  
102 bound to geological formations which were buried, then exhumed and exposed fortuitously at  
103 the surface in landscapes where they could be accessed, tested and exploited if suitable. On  
104 the other hand, silcretes are directly linked to the landscapes in which prehistoric people lived.  
105 Those silcretes that frame and armour landsurfaces as duricrusts and lags were highly visible  
106 and available for testing and exploitation over wide areas of continents.

107        The most widely known silcrete duricrusts occur now in the desertic landscapes of  
108 inland Australia, South Africa, Saharan Africa and the northeast of Brazil. They armour  
109 plateaux and mesas dissected by erosion, thus maintaining steep scarps or ‘breakaways’ cut in  
110 vari-coloured soft substrate formations that confer a spectacular aspect to the lands that  
111 fascinated prehistoric people and still fascinate tourists. Silcretes are also important regolith  
112 features and frame a number of landscapes in temperate regions (for example the  
113 superimposed plateaux in the Paris Basin), but are less spectacular because they tend to be  
114 covered and hidden by vegetation. During the glacial periods in the Late Cenozoic these  
115 northern countries were somewhat bare of vegetation, with contrasting relief.

116        Although there are several forms of silcrete that can be found and recognised on many  
117 continents, only some appear to have been suitable for stone tool making. The sites that have  
118 generated starting materials for appropriate tools can be readily recognised in the field from  
119 the litter of flakes and chips discarded during tool production. Mineralogical and  
120 micromorphological properties can explain physical appearance, hardness, lustre and potential  
121 durability of silcrete tools and, in some cases, in some regions, point to their provenance.  
122 However, archaeologists may not be particularly focussed on sources of silcrete (or other  
123 forms of siliceous rocks) that have been opportunistically exploited for the production of

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124 stone tools, but rather seek to recognise specific source materials that have been valued from a  
125 technological, aesthetic or symbolic viewpoint and which potentially have been transported  
126 long distances. Identifying provenance from this perspective permits them to potentially trace  
127 out migration patterns, exchanges, and possibly social behaviour.

## 128 **2 Behaviour of silica in regolith environments**

129 In order to explain the internal structures of silcretes, as well as their distribution and  
130 location in specific landscape settings, it is important to understand some concepts of silica  
131 behaviour in surficial environments.

### 132 *2.1 Silica solubility*

133 The solubility of amorphous silica in water under earth-surface conditions is about 120  
134 mg/L SiO<sub>2</sub> while the solubility of all other silica phases is significantly lower than this (Fig.  
135 1). The most soluble phases in a geochemical system control the precipitation of the less  
136 soluble ones and, in this way, amorphous silica in equilibrium with a solution is able to  
137 sustain the crystallisation of every other crystalline silica phase. In turn, cristobalite,  
138 tridymite, chalcedony and opal in equilibrium with a solution are able to sustain the  
139 crystallisation of quartz (Fig. 1; Garcia-Hernandez, 1981; Gislason *et al.*, 1993; Iler, 1979;  
140 Siffert, 1967).

141 Detailed mineralogical and petrographic studies of silcretes show a consistent and  
142 specific sequence of transformation/recrystallisation from more to less soluble phases,  
143 namely:

144 opal → microcrystalline quartz → euhedral quartz

145

146 These transformations are the mainspring of the evolution of silcrete profiles and  
147 especially account for the development of complex nodular structures in some forms. The  
148 succession of recrystallization is irreversible and ultimately favours the formation of quartz  
149 (Thiry and Millot, 1987).

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150           It should be noted that most natural groundwaters around the world have a silica  
151 content between 12 -18 ppm SiO<sub>2</sub> (Garrels and Christ, 1965, Swanberg and Morgan, 1978;  
152 White *et al.*, 1963) that is roughly in equilibrium with clay minerals. This means that most  
153 groundwaters are oversaturated with respect to quartz (solubility 4 - 7 mg/L) and are thus  
154 potentially able to precipitate quartz when favourable physico-chemical conditions occur. As  
155 pointed out by Thiry *et al.* (2014), solutions with a high silica content favour the formation of  
156 crystal nuclei and crystal defects that restrict the growth of crystals and consequently the  
157 precipitation of amorphous or low crystallinity silica varieties. However, in dilute solutions,  
158 the number of nuclei and impurities remains limited. As a consequence, pure silica crystallites  
159 generally form and large quartz crystals can develop or, under less favourable conditions,  
160 microcrystalline or fibrous silica varieties are formed.

161           If silica is present in solution it has to be precipitated in order to generate silicification  
162 and ultimately produce a silcrete. Several tracks are possible. It has long been thought that  
163 increasing the concentration of the solution by evaporation was the principal triggering factor  
164 for silica precipitation in relation to silcrete formation (Auzel and Cailleux, 1949; Millot  
165 1970; Smale, 1973; Storz, 1928). This hypothesis resulted mainly from studies of the  
166 widespread silcrete duricrusts in current desertic environments, for example in Australia,  
167 South Africa and North Africa. It has now been established that these silcretes do not relate  
168 to present day landscapes and climates and are in fact much older. Moreover, it is unlikely  
169 that evaporation could be effective in developing meter-thick silicified horizons because it  
170 would be much reduced, or cease, as soon as silica cementation commenced. Nevertheless,  
171 evaporative processes may assist in concentrating silica in solution in near-surface  
172 environments.

173           In contrast to evaporation, silica solubility decreases exponentially with decreasing  
174 temperature. Between 12.5 and 0°C, the solubility of quartz decreases to half its initial value  
175 (Fig. 2). This appears to be a very effective factor in precipitating silica from solution: it had  
176 never been envisaged as a mechanism for precipitating silica in the supergene realm until  
177 proposed by Thiry *et al.* (2013, 2015b) but was commonly advocated in hydrothermal  
178 environments.

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179           Increasing the concentrations of salts in solution is another efficient way to decrease  
180 silica solubility: the solubility of amorphous silica decreases with increasing salt  
181 concentration to saturation (Fig. 3), up to 96% in the case of CaCl<sub>2</sub> and up to 30% in the case  
182 of a NaCl saturated solution (Marshall, 1980). The mixing of a silica solution with chlorite-  
183 and/or sulphate-rich brine is certainly a very effective mechanism for inducing the  
184 precipitation of silica. The often advocated relation between silicification and warm and dry  
185 climates most probably results from the negative relationship between increasing  
186 concentrations of evaporite brines and silica solubility.

## 187           2.2 Key geochemical processes in regolith solutions

188           As silica in regolith solutions mainly originates from the weathering or alteration of  
189 alumino-silicates (such as feldspars and clay minerals) in rocks and sediments, the relative  
190 solubilities of Si and Al must be taken into account. Si and Al have different domains of  
191 stability (Fig. 4).

192           (1) In acidic environments, Al and Si have contradictory behaviours. Al is more  
193 soluble than Si, and so alumino-silicates (clays and feldspars) are destroyed and leached of Al  
194 whereas Si in the silicate frameworks, and silica minerals, remain essentially *in situ*, most  
195 often in the form of opal;

196           (2) At neutral pH, Al and Si behave similarly. Al and Si both have a low  
197 solubility. Both silicates and silica are relatively stable in this domain;

198           (3) In alkaline environments Al and Si are both highly soluble. Silicates and silica  
199 may be altered, but such environments are rare and restricted in continental landscapes (for  
200 example alkaline lakes in the east African graben).

201           Highly acidic conditions appear to be especially favourable for the degradation of  
202 silicates and clay minerals via dissolution and leaching of Al and other cations to concentrate  
203 silica. However, such acidic environments are not common in regolith environments and  
204 require specific conditions to develop.



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230 in solution to infiltrate, and initiate silicification of, specific zones or horizons in the regolith  
231 (*absolute* accumulation of silica).

### 232       2.3 *Silicification in the regolith*

233           In general, silcretes have formed as a result of either *absolute* or *relative* accumulation  
234 of silica.

235           *Relative* accumulation of silica occurs primarily as the result of the dissolution and  
236 leaching of alumino-silicate minerals, specifically silicates such as feldspars and clays (Rayot  
237 *et al.*, 1992; Thiry *et al.*, 2006). The process requires an acidic environment that is aggressive  
238 towards these minerals and generates the loss of non-Si elements, resulting in an *in situ*  
239 concentration of silica. There appears to be no significant transport of Si within the system  
240 but only limited local reorganization and adjustment. However, leaching of cations other than  
241 silica generates a significant loss of material leading to the disruption and collapse of primary  
242 fabrics and structures.

243           *Absolute* accumulation occurs through importation of silica to the system. The silica  
244 may replace pre-existing minerals or simply fill voids, fractures and intergranular pores. The  
245 silica has to originate from somewhere else and has to be brought into the horizon that  
246 undergoes silicification. With regard to the relatively low solubility of silica in surficial  
247 waters and the relatively large volume of silica needed to infill pore spaces to effect  
248 silicification, this kind of silica accumulation requires large volumes of water to flow through  
249 the horizon. This is only conceivable in groundwater outflow zones (springs, seeps) and  
250 implies landscape incision in order to intersect groundwater tables. Primary geological  
251 structures in the host horizon are characteristically preserved.

## 252   **3       Varieties of silcrete**

253           Silcretes take a wide variety of forms and detailed investigations of these forms from  
254 the field scale to that available via scanning microscopy provide clues to their origin. Aspects  
255 such as field distribution, geomorphological patterns and relationships to current and former  
256 landscapes, petrology and mineralogical-geochemical characteristics have all to be

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257 considered. We group silcretes into two classes: groundwater silcretes that form mainly as a  
258 result of the absolute silica accumulation of silica in regolith environments; and pedogenic  
259 silcretes that reflect the relative accumulation of silica, as well as a complex of dissolution,  
260 eluviation and illuviation processes typical of soil environments. Nash and Ulliyott (2007)  
261 have suggested a sub-classification of ‘*non-pedogenic*’ silcretes into ‘*drainage-line*’ and  
262 ‘*pan/lacustrine*’ types’, but these forms can be encompassed satisfactorily within groundwater  
263 silcretes from our perspective. Of particular interest in the landscape context is that volume is  
264 mostly conserved during the formation of groundwater silcretes, whereas volume is  
265 substantially lost during the formation of pedogenic silcretes, some of which can be metres  
266 thick in profile.

### 267 3.1 Groundwater silcretes

268 The main characteristics of groundwater silcretes are their disposition as a series of  
269 superposed silicified lenses in the regolith and the preservation of primary host-rock  
270 structures (stratification, bioturbation, fossils). There are two main varieties: those that occur  
271 in sands and develop into massive quartzitic facies by infilling and cementation of pore spaces  
272 between detrital quartz grains; and those that are created by epigenetic replacement of  
273 limestones, claystones and even gypsum horizons, and therefore have more irregular and  
274 discontinuous shapes.

#### 275 3.1.1 Quartzitic types

276 These are typically formed in sands by cementation of the detrital quartz grains via  
277 syntaxial quartz overgrowths, or other forms of silica, precipitated in pore spaces. The initial  
278 condition is always a porous sediment. Quartzitic silcretes generally form well-confined flat-  
279 lying pans within unconsolidated or poorly cemented sands. Those in the Fontainebleau Sand  
280 have been studied in detail and are good examples.

281 In the Paris Basin, flat-lying quartzitic silcrete lenses (1-8m thick) occur at different  
282 levels within the Fontainebleau Sand formation (Thiry *et al.*, 1988; Thiry *et al.*, 2013; Thiry *et*  
283 *al.*, 2015a). The silcrete lenses typically crop out on the edges of plateaux or in valley  
284 margins but pinch out rapidly under the limestone-capped plateaux, as shown by drill holes

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285 and observations in sand quarries (Fig. 6). This strong link between silcrete distribution and  
286 present-day geomorphology suggest that these silcretes developed relatively recently in and  
287 near outcrop zones of the Fontainebleau Sand. Sand calcite contained in the silcretes date the  
288 silicification to Pleistocene glacial periods (Thiry *et. al.*, 2013).

289         Geomorphological and geological constraints suggest that silicification occurred in  
290 groundwater outflow zones (springs, seeps) during landscape incision (Thiry *et al.*, 1988;  
291 Fig. 7). The key process sequence is:

- 292         • precipitation of silica in horizons of the Fontainebleau Sand in a zone of groundwater  
293             outflow; and
- 294         • resumption of erosion leading to lowering of the groundwater table and the formation of  
295             a new silcrete pan via groundwater outflow during a subsequent period of landscape  
296             stability;

297 The superposition of separate silcrete pans rather than the formation of a continuous zone of  
298 silicification reflects periodicity in valley down-cutting and landscape stability.

299         The silica necessary for cementation of the Fontainebleau Sand to form quartzitic  
300 silcrete has come from groundwater contained within the formation. Its silica content ranges  
301 from 10 - 15 ppm, which is about three times the saturation value of quartz (4 - 6 ppm) at  
302 surface temperatures and pressures.

303         The correlation of silica cementation with glacial periods, as determined by age dating  
304 of contained calcite crystals, suggests that silica precipitation occurred by cooling of  
305 groundwaters along discharge pathways towards outflow zones in frozen soils in the valleys.  
306 Hydraulic and geochemical mass balance calculations, based on the silica content of water in  
307 an average current-day spring, indicate that cementation could potentially take place over  
308 about 2 000 years. This is a short geological time span and totally changes previous  
309 interpretations of the time-scale for the formation of some silcretes.

310         Similar quartzitic silcretes appear to have formed under cold environments during the  
311 Pleistocene in North America (Ludwig and Paces, 2002; McCoy, 2011), and such an origin  
312 may apply to other occurrences, like those within white sandstone formations in Germany

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313 (Götze and Walther, 1995) and in the Czech Republic (Mikulas, 2002), Greece (Skarpelis,  
314 2006), southern France (Parron *et al.*, 1976) and Spain (Bustillo and Bustillo, 2000; Parcerisa  
315 *et al.*, 2001), for which the origins have not yet been considered in detail. Quartzitic silcrete  
316 pans may also develop in very different environments, including those that have not  
317 apparently experienced cold climates. For example, in Australia (Thiry and Milnes, 1991)  
318 and southern Africa (Summerfield, 1983; Nash *et al.*, 1998), quartzitic silcretes may have  
319 formed in groundwater environments in dry paleoclimates where silica precipitation could be  
320 attributed to a mixing of fresh and saline waters. Nevertheless, all examples relate to  
321 groundwater outflows in incised landscapes and for which silica has been imported in order to  
322 provision precipitation and cementation.

### 323 **3.1.2 Silicified limestone/calcrete types**

324 Many lacustrine limestone formations contain irregular silicified masses and have long  
325 been described from dry regions such as the Kalahari Desert, North Africa and Australia, as  
326 well as in more humid countries like France, Spain and the USA (Alimen and Deicha, 1959;  
327 Banks, 1970; Daley, 1989; Kaiser, 1928; Nash *et al.*, 2004; Thiry and Ben Brahim, 1997).  
328 The silica was generally thought to have precipitated during dry periods at the time of  
329 limestone deposition in confined environments. However, petrographic observations and  
330 geochemical data have generated new ideas about the formation of these silcretes.

331 The extensive Tertiary lacustrine limestones of the Paris Basin form superimposed  
332 plateaux that constitute the dominant geomorphologic features. Typical examples are the Brie  
333 and Beauce plateaux. Almost all of these limestones contain zones of silicification (Cayeux,  
334 1929). Their size varies from millimetre-sized dots to siliceous bodies several tens of meters  
335 long. The silicified facies have an irregular distribution (Fig. 8): locally, they can form over  
336 10% of a quarry cliff-face, but are absent a short distance away. On the regional scale they  
337 average about 5% of the volume of the lacustrine limestone in the southern Paris Basin.

338 Two types of silicification coexist: deposits of quartz and chalcedony in voids; and  
339 epigenetic replacement of the limestone matrix by microcrystalline quartz, with preservation  
340 of the primary sedimentary structures. Thin sections show a systematic link between

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341 silicification and zones of high porosity, either preserved or partly infilled by silica deposits  
342 (Thiry and Ribet, 1999).

343           Because the silicified limestones are pure, without any sandy or clayey layers, silica  
344 had to be imported from elsewhere. And as the limestones have never been deeply buried, the  
345 importation of silica can only have been via substantial near-surface groundwater flows,  
346 which could explain the relationship between porosity and the intensity and extent of the  
347 silicification. This is a very different mechanism of silica accumulation from that represented  
348 by flints and cherts. In this case, silicification occurs during burial diagenesis whereby there  
349 is epigenetic replacement of limestone by silica originating in microfossils in specific  
350 sedimentary layers, without the formation of any voids.

351           Substantial groundwater outflows are only possible after uplift and incision of the  
352 limestone formation. In the Paris Basin, such conditions were only fulfilled during the  
353 Pliocene and the Quaternary periods and, therefore, the silicification had to occur relatively  
354 quickly (Thiry and Ribet, 1999). Similar groundwater silicification of limestone has already  
355 been described in present day hydrologic systems in inland Australia (Arakel *et al.*, 1989;  
356 Benbow, 1993), southern Africa (Nash *et al.*, 2004) and Spain (Armenteros *et al.*, 1995). As  
357 for quartzitic silcretes, silica precipitation was probably triggered by different geochemical  
358 mechanisms (water cooling or water mixing, or others), under very different climatic and  
359 environmental conditions. However, the resulting petrographic fabrics are similar: dissolution  
360 of calcite making space for precipitation of silica, which may lead to very contrasted fabrics.

361           Weathering of Paris Basin silicified lacustrine limestone during Pliocene-Quaternary  
362 times produced cavernous silicifications called "meulières" which delineate plateau surfaces  
363 in the southern Paris Basin (Beauce Plateaux; Thiry, 1999). The meulières show a large  
364 variety of facies from massive to highly porous 'cellular' facies and can be weathered and  
365 desilicified into friable blocks, even breaking down into a rough sand-like material.

### 366           **3.1.3 Porcellanite/jasper types**

367           Fine grained silcretes, obviously resulting from silicification of primary clay-rich  
368 materials, form specific facies, generally of glassy appearance, and often resembling flint or  
369 chert except for their colour which is generally due to iron-oxide inclusions. Such materials

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370 have been described in several geological contexts (Boule, 1888; Millot *et al.*, 1959, Valleron,  
371 1981).

372           Such silicifications are widespread in inland Australia and are commonly associated  
373 with bleached and weathered profiles in rocks and sediments ranging in age from Precambrian  
374 to Quaternary (Simon-Coinçon *et al.*, 1996). The profiles are up to 60m deep. Their main  
375 characteristics are white or pastel colour and low bulk density, together with the occurrence of  
376 alunite, iron oxide mottling, extensive bioturbation by termite burrows that penetrate to great  
377 depth, and ubiquitous gypsum that has crystallised in fractures and voids (Thiry *et al.*, 1995;  
378 Thiry *et al.*, 2006). The bleached material is mainly formed of quartz, large booklet-like  
379 kaolinite and opal A, with alunite and gypsum in pods and veins (Fig. 9). In some localities  
380 the burrows and their fillings, as well as the enclosing saprolite, have been silicified. K/Ar  
381 dating of alunite in these profiles indicates a Miocene age for the bleaching and weathering,  
382 varying from about 16 - 18 Ma in the Coober Pedy opal field to about 8 - 12 Ma in the  
383 Andamooka opal field (Bird *et al.*, 1990).

384           These profiles have resulted from alteration under acidic conditions in a saline  
385 groundwater environment nearly saturated in gypsum (Thiry *et al.*, 1995). The bleaching was  
386 related to the lowering of the groundwater table. Arid conditions at the time restricted water  
387 flow and allowed an acidic environment to develop to leach original alumino-silicates, retain  
388 Si (as amorphous silica and quartz), but not completely lose some of the most soluble  
389 elements like K, Ca, SO<sub>4</sub> and Al from the system that form nodules and veins of alunite and  
390 gypsum in places. Elevated concentrations of anatase (Fig. 9) could indicate that Ti released  
391 from primary alumino-silicates has also been retained.

392           Horizontally disposed lenses or pods of porcellanite or jasper subsequently developed  
393 in the bleached saprolite (Fig. 9). Primary sedimentary structures and fabrics, as well as some  
394 fabrics relating to weathering and bleaching, in particular iron-mottling, have been retained  
395 within these silica-indurated horizons. The superposition of several silica-indurated layers  
396 relates to still-stand positions of paleo-water tables which rose significantly after the  
397 widespread bleaching, thus reflecting a return to more humid climatic conditions.

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398 Low concentrations of clays in the porcellanite and jasper horizons compared with the  
399 host regolith (Fig. 9) points to acidic environments that probably developed via ferrolysis and  
400 generated silica via relative accumulation. However, micromorphological examination shows  
401 that the silicified horizons are cemented by various silica phases. The silica cements were  
402 precipitated from water inflow, either by infiltration through the upper part of the profile, or  
403 by lateral flow of groundwater containing silica through the profile. The silica was probably  
404 derived from dissolution of silica (opal-A, quartz) and silicates (clays) in the bleached profile.  
405 The formation of these groundwater silcretes required a consistent hydraulic flow regime that  
406 was probably related to down cutting of the landscape (Simon-Coinçon *et al.*, 1996). Silica  
407 precipitation points to mixing of fresh silica-charged water with sulphate-rich groundwater,  
408 thus lowering silica solubility.

### 409 3.2 Pedogenic silcretes

410 Pedogenic silcretes developed near the landsurface, within soil environments, and  
411 display typical soil structures (geopetal features) related to infiltration of downward  
412 percolating water (such as differentiated horizons, eluviation and illuviation structures).  
413 Details of this silcrete type come mainly from Eocene occurrences in the Paris Basin (Thiry,  
414 1981). Specific characteristics and mechanisms of formation are also derived from  
415 observations of similar materials in Australia (Milnes and Thiry, 1992; Simon-Coinçon *et al.*,  
416 1996; Thiry and Milnes, 1991) and Central America (Elsass *et al.*, 2000).

417 In the southern part of the Paris Basin, silicified pans and lenses cap Eocene detrital  
418 kaolinitic deposits. These are very hard, tightly cemented quartzitic silcretes that break up into  
419 variably sized blocks, with puffy, tear-like shapes, coated with deposits ('cappings') of  
420 yellow-white opaline silica that mask the internal structures. Typically, the silcretes have a  
421 columnar structure with characteristic laminated cappings on the columns (Fig 10). They are  
422 about 2 m thick and display several distinct horizons with systematic micromorphological and  
423 mineralogical structures (Thiry, 1981).

424 A lower granular horizon consists of a sandy claystone with millimetric- to  
425 centimetric-sized granules of microcrystalline quartz and opal. TEM and electron diffraction

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426 studies of the clay matrix show that hexagonal-shaped kaolinites have corrosion embayments  
427 and are coated and welded together by a silica gel.

428           An overlying columnar horizon has a matrix composed of detrital quartz grains with  
429 irregular overgrowth apophyses which grade into titania-enriched microcrystalline quartz.  
430 Ghosts of granules are easily recognizable within the matrix. Illuviation cutans of opal have  
431 developed in fractures. The youngest laminae, at the top of the illuviation structures, always  
432 consist of opal with a low titania content. The initial laminae, at the base of the illuviation  
433 structures, commonly have a nodular structure and are formed of microcrystalline quartz with  
434 a higher titania content. This sequence indicates a progressive recrystallisation of the opal.  
435 Recrystallisation is accompanied by loss of silica, which induces the destruction of the  
436 primary structures and relative accumulation of titania.

437           A more massive horizon, with complex nodular and pseudo-breccia fabric, is found at  
438 the top of the profile. It is devoid of opal and the microcrystalline quartz matrix has partly  
439 dissolved. Titania has accumulated in rims around the nodules. Euhedral quartz has developed  
440 in the voids and some quartz grains show overgrowths.

441           Pedogenic silcretes occur widely in central and southern Australia and have been  
442 described in detail from the opal fields (for example Fig. 9, near-surface horizon) and the  
443 region around Alice Springs (Milnes and Thiry, 1992; Milnes and Twidale, 1983; Simon-  
444 Coinçon *et al.*, 1996; Thiry and Milnes, 1991). Their morphology is typically that described  
445 by Thiry and colleagues from around the Paris Basin (Barrois, 1878; Thiry, 1981), with wide  
446 variations in the appearance of the columnar facies being evident in different localities. As  
447 well, pedogenic silcretes occur widely in north-western Europe, in Belgium on the Ardennes  
448 (Gosselet, 1888), in Germany on the Slate Mountains massifs (Lange; 1912; Teichmüller,  
449 1958) and in the London Basin (Kerr, 1955, Summerfield, 1980). They occur in South African  
450 and Botswanan landscapes where Summerfield (1983) described highly variable textures and  
451 fabrics classifying the main petrographic characteristics as '*grain-supported fabric*' (quartzitic  
452 silcrete with quartz overgrowths), '*floating fabric*' (silicified clayey matrix with coarse  
453 detrital grains), and '*glaebular fabric*' (containing illuviation structures typical of soils).

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454 In terms of the origin of pedogenic silcretes, opal initially formed in the granular  
455 horizon and at the base of joints in the columnar horizon where water circulation was slow  
456 and stagnation had occurred. Higher in the profile, the microcrystalline matrix dissolved.  
457 Well-crystallised quartz crystals formed at the top of the profile. Profiles clearly show a  
458 migration of silica from top to bottom and the inheritance, in the uppermost horizons, of  
459 micromorphological features which initially developed in the lowermost horizons. This  
460 inheritance demonstrates that silicification progressively ‘eats’ into the landscape like a  
461 weathering front. The greatest amount of silica comes from dissolution at the top of the  
462 profile followed, from top to bottom, by a sequence of precipitation and re-dissolution events.

463 The close link between leached and confined environments does not imply a strict  
464 synchronism of degradation and construction. The two systems work in an alternating fashion,  
465 whereas periods of loss (leaching) and accumulation (precipitation) follow one another more  
466 or less sequentially.

467 Initially silica originated from the degradation of clay minerals at the base of the  
468 profile, possibly in a highly acidic environment triggered by ferrollysis. The mineral sequence,  
469 from top to base of the profile, results from a progressive concentration of silica and other  
470 cations in infiltrating and downward-moving solutions. The silica content of the solution  
471 could be concentrated by 2 to 4 times through evaporation during dry periods and silica  
472 precipitation favoured by increasing cation concentration.

#### 473 **4 Characteristics of silcretes of relevance to tool making**

474 Silcretes show numerous variations in their morphology and their spatial arrangement  
475 but also vary in their composition and internal texture. A classification of silcretes based on  
476 micromorphological fabrics by Summerfield (1983) has been used widely in archaeological  
477 studies to identify different types of silcrete artefacts. Indeed, each silcrete type may display  
478 various types of cement and porosity, with sometimes decimetric scale variation, but this does  
479 not relate to their convenience or value for tool making. Particular facies, with interesting  
480 mechanical properties or appearance could exist within larger masses and may have been  
481 specifically sought and used.

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482           Various crystallization fabrics have developed according to the nature of the host  
483 formation and/or the chemistry of the silica-bearing solutions. The nature of silica cements  
484 and the geometrical relations between crystals are the predominant determinants of the  
485 mechanical properties of silcretes. Substantial research in this field has been undertaken in  
486 Australia from an archaeological perspective (e.g. Domanski *et al.*, 1994; Webb and  
487 Domanski, 2008). These investigations have determined that tool-making depends on two  
488 main characteristics: the hardness of the material to form cutting and wear-resistant tools, and  
489 the capacity to produce a regularly curved conchoidal fracture that permits the detachment of  
490 long and thin knappings.

491           It is not within our capability to examine the knappability of silcrete types, nor to  
492 make reference to the variety of silcrete facies used by prehistoric peoples to make stone  
493 tools. Much of the formal understanding of the mechanical properties of silcretes and other  
494 rocks from which stone tools were made, and the influence on these properties by heat  
495 treatment, which is believed to have been commonly practiced, has been comprehensively  
496 detailed (e.g. Domanski *et al.*, 1994; Domanski and Webb, 1992; Domanski and Webb, 2007;  
497 Webb and Domanski, 2008). Our objective is to illustrate how some petrographic fabrics of  
498 silcretes, which we use to help unravel their origin and environment of formation, may effect  
499 knappability.

500           Broadly speaking, silcretes that have the finest grain size and the least porosity display  
501 the best developed conchoidal, vitreous fracture surfaces. From a petrological and  
502 mineralogical perspective, the finest grain size will occur in those silcretes formed  
503 predominantly of opal, which can be a complex of hydrated, poorly crystallised silica phases  
504 (opal-A, opal-CT) and is consequently comparatively soft (Mohs hardness ~5-6). In those  
505 silcretes in which the matrix is dominantly microcrystalline quartz, of which there are several  
506 types ranging from granular to fibrous varieties, the Mohs hardness is that of quartz (7) and,  
507 depending on the microporosity, fracture surfaces can be glassy in appearance. As the  
508 abundance of detrital framework quartz grains increases, for example in pedogenic silcretes  
509 and some quartzitic groundwater silcretes, the smoothness and lustre of fracture surfaces is

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510 controlled essentially by the bonding between framework grains and matrix silica, and can be  
511 influenced by the abundance of other constituents including micro-cryptocrystalline anatase.

#### 512 4.1 *'Sandstone' cementation*

513 In clean sands, without a clay matrix, such as the Fontainebleau Sand, precipitated  
514 silica builds on the crystal structure of each detrital quartz grain, leading to the development  
515 of euhedral crystal faces. On continued precipitation, crystal faces come into contact, and  
516 finally form triple point junctions at the site where the pore space has closed (Fig. 11). The  
517 overgrown and expanded quartz grains are not welded together, but only meshed in a compact  
518 way. This provides cohesion and hardness to a quartzitic silcrete formed in this manner.  
519 When broken by hammering or knapping, the fractures can cross cut quartz grains but tends to  
520 follow the euhedral crystal faces of the overgrowth quartz that make contact between the  
521 grains. This results in a rough fracture surface of sugary appearance. Such fractures are not  
522 suitable for making thin blades or sharp cutting edges.

523 Cementation of quartz grains may also be achieved as the result of precipitation of  
524 silica in pore spaces between the grains (Fig. 11), either in the form of opal (quasi-amorphous  
525 silica) which may have later re-crystallized into chalcedonite, or direct precipitation of  
526 chalcedonite sheaves. These silica deposits give rise to stronger cementation than quartz  
527 overgrowths and this is linked to the bonding between the detrital quartz grains and the silica  
528 precipitates and the complex intercrystallite connections in the pore spaces. Accordingly, the  
529 fracture in these silcretes tends to be 'clean' and cuts across the detrital quartz grains. When  
530 the detrital quartz grains are relatively small and the silica deposits occupy the main part of  
531 the porosity, the fractures may even be conchoidal and lustrous. This is most likely when the  
532 pore fillings are of microcrystalline quartz.

#### 533 4.2 *Silicified limestones*

534 In silicified limestones the carbonate matrix has been replaced by silica. The  
535 substitution is mainly epigenetic, which means that primary limestone structures such as  
536 nodules and fossils are replaced by silica and conserved. For this to occur, there can be no

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537 dissolution of the carbonate before replacement: substitution occurs step by step along a clear  
538 'alteration front' between existing limestone and incoming silica. This replacement implies  
539 concomitant leaching of the carbonate and precipitation of silica. Voids are necessary for  
540 water circulation and their shape and size determines the density and 'quality' of the  
541 silicification.

542         If the voids have the form of micro-karst, which develops as the silica precipitates  
543 (Fig. 12), the resulting silcrete could be very dense, as translucent as flint and with a lustrous  
544 conchoidal fracture. On the other hand, when the dissolution features are larger, or when  
545 carbonate residues remain as impurities in the silicified zones, then the resulting silcrete has a  
546 dull aspect and a rough surface and is much less convenient for tool making.

547         Quartz is generally the main silica mineral and occurs as both epigenetic replacement  
548 and also euhedral overgrowths in pores. Some of the fine microcrystalline matrix may result  
549 from recrystallization of primary opal. Chalcedonite and laminated opal deposits occur in  
550 pores in some sites (Fig. 12). The presence of opal and poorly crystallized forms of silica  
551 may be favourable for heating transformation of these silcretes which is sometimes observed  
552 naturally in outcrop in Australia.

### 553         4.3 *Porcellanites and jaspers*

554         These materials are of fine grain size and are formed mainly of various petrographic  
555 varieties of opal. The matrix resulting from the alteration of the primary material is generally  
556 formed of milky opal made opaque by impurities and microporosity. The most compact  
557 samples, with a glossy break, also contain translucent concretionary opal that cements the  
558 pores within the matrix (Fig. 13) and this may, in some samples, make up more than 50 % of  
559 the silcrete. Concretions generally show successive sequences of silica precipitation in which  
560 there are alternations of thin laminae and thicker botryoidal layers of clear and brown opal.  
561 The opal has been deposited uniformly around the voids, indicating a saturated groundwater  
562 environment.

563         Crystalline varieties of silica occur also in these silcretes. They have formed by  
564 recrystallisation of opal, either preferentially in some concretions as part of the sequence of

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565 deposition, or as in response to some form of alteration front (Fig. 13). The initial opal  
566 deposits have often recrystallized to chalcedonite while the brown opal of the matrix has  
567 transformed to microcrystalline quartz.

568         The opal deposits generally strengthen the silcrete and generate lustrous conchoidal  
569 fractures when knapped. However, when exposed at the landsurface in Australia,  
570 porcellanites can exhibit extensive crazing as a result of volume changes on dehydration of  
571 the opal phases.

#### 572         4.4 *Pedogenic silcretes*

573         Pedogenic silcretes are highly variable in form and facies which results from the  
574 diversity of the silicified parent materials (colluvium, clayey sand, clay silt, granite, bedrock  
575 sandstone) and the position of the sample within the profile (columnar or nodular facies).  
576 Adding to the variability is the mineralogical composition which records successive  
577 recrystallization stages within the profile.

578         The most abundant and the most typical facies are tightly indurated by a dominantly  
579 microcrystalline quartz cement and have a conchoidal and lustrous fracture. Opal is mostly  
580 limited to less indurated facies at the base of the profiles and to specific micromorphological  
581 features within the profile that are always limited spatially and in volume.

582         The density and homogeneity of the microcrystalline quartz matrix can result in a  
583 material that is well-suited for knapping. The amoeboid microcrystals of quartz in net-like  
584 arrangement (Fig. 14) confers a structure and texture that mimics the characteristics of some  
585 flints. Silicified silty claystones that contain fine-grained detrital quartz and a substantial  
586 microcrystalline matrix can be knapped particularly well.

587         A principal criterion for recognising pedogenic silcretes is the omnipresence of  
588 illuviation structures, particularly cutans at the base of voids and cappings over quartz grains  
589 and granules. These are very clearly distinguished because they are outlined by opaque  
590 microcrystalline titania (Fig. 14). As well, the common shard-like remnants of detrital quartz

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591 grains point to significant dissolution and concomitant ‘repacking’ of the framework prior to  
592 cementation, leading to a net volume loss in the profile as a whole.

## 593 **5 Silcrete in landscapes**

594 There are two aspects relating to silcretes in a landscape context. In the first case,  
595 from the geological perspective, the occurrence and form of silcretes, particularly where  
596 profile observations can be made, provides the basis for interpreting the environment of  
597 formation of the silcrete. Macroscopic form and structure indicate the relative simplicity or  
598 complexity of the processes of formation, point to single stage or overprinting characteristics,  
599 and indicate conservation of volume and preservation of fabric and structure (groundwater  
600 environments) or dissolution and loss of material with consequent and complex disruption in  
601 the profile and, presumably, of the landsurface (pedogenic silcretes).

602 In the second case, within each category of silcrete, and even within a given category  
603 in a particular location, the macroscopic and microscopic fabric and structure can vary  
604 considerably. This is less so in the case of groundwater silcretes than for pedogenic silcretes.  
605 Only particular forms of silcrete appear to have had value for the production of stone tools.  
606 Mulvaney and Kamminga (1999), for example, point to the possibility that some forms of  
607 silcrete could have been selected for tool production for aesthetic or symbolic reasons,  
608 including their colour, which can be variable. Holdaway *et al.* (2008) and others (Doelman *et*  
609 *al.*, 2001) make the distinction between ‘outcrop silcrete’ and the extensive lag gravels  
610 (‘gibber plains’), the latter (also called ‘stony deserts’) resulting from downwasting of the  
611 ancient pedogenic silcrete-capped landsurfaces and associated groundwater silcrete-  
612 impregnated regolith over large areas of inland Australia (Fujioka *et al.*, 2005). They point  
613 out that artefacts were made from each source, but that outcrops were more difficult for  
614 Aboriginal people to exploit for tools than gibber lags because of the greater supply of hand-  
615 sized cobbles in the latter.

616 Thus, if we want to explore the use of silcrete as a raw material for stone tools, for  
617 example to map the routes of particular source materials and thus delimit territories and  
618 exchanges between prehistoric groups of peoples, it is necessary to locate these materials in

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619 the landscapes of the studied areas and the territories of the people exploiting them. In this  
620 context, inventories of the locations of specific silicified formations, and their petrographic  
621 characteristics, would be required for both local and regional areas. This research would  
622 utilise an understanding of silcretes and silcrete facies in a landscape context because  
623 prehistoric peoples had deeply-embedded connections with landscapes and their features.

### 624 5.1 *Inland Australia*

625 As is well known, pedogenic silcretes in Australia armour glacis (pediments) on the  
626 edges of plateaux and around paleorelief (Milnes and Thiry, 1992; Milnes and Twidale, 1983;  
627 Simon-Coinçon *et al.*, 1996; Thiry *et al.*, 1991; Twidale and Milnes, 1983a, b). At the edges  
628 of scarps extending from the Arcoona Plateau in South Australia, for example, silicification is  
629 restricted to thin ‘skins’, with a very high titania content, coating joint fractures and other  
630 surfaces on quartzite bedrock (Hutton *et al.*, 1972; Fig. 15). On the proximal part of the Beda  
631 pediment, just downslope from the scarp-foot zone of the plateau, silcrete is more extensive  
632 and almost only formed of thick cappings (5 to 20 cm thick) on bouldery quartzite scree,  
633 whereas in the distal areas of the pediment silcrete forms a regular horizon with columnar  
634 structure within the pediment. Thicker silcretes occur in the transition zones between the  
635 glacis and the lowland plains, where water discharge is still important but flow rates have  
636 slowed.

637 The facies with thick cappings on scree boulders is symptomatic of the complexity  
638 that can face any archaeological search for the source of silcrete tools. These cappings are of  
639 very hard silcrete with a convex lustrous break and have been exploited for tools as shown by  
640 the litter of flakes and chips surrounding the outcrops. They may have been sought  
641 specifically because of certain characteristics. However, this particular facies is of very  
642 limited extent and locating it as a source of particular stone tools would only be possible after  
643 a very detailed inventory of a wide area.

644 In localities along the scarp of the Stuart Range, near Coober Pedy, in northern South  
645 Australia, a comprehensive analysis of the geomorphology of the region (Simon-Coinçon *et*  
646 *al.*, 1996) delineated the age relationships between the different weathering and silicification

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647 features (Fig. 16). The pedogenic silcrete armours a wide paleopediment (tableland) dipping  
648 from the Stuart Range near Lake Eyre to the Eucla Basin in the southwest, a distance of about  
649 500 km. The weathered and bleached profiles are younger. Groundwater silcrettes,  
650 specifically quartzite and diverse porcellanite horizons, post-date the bleaching. Downcutting  
651 and erosion ultimately generated the breakaway scarps now characteristic of the region.

652         The large variety of silicified materials in the Stuart Range landscapes are shown  
653 schematically in more detail in Figure 16. Pedogenic silcrettes may be locally buried beneath  
654 younger deposits (clayey sediments and red-brown hardpans) but are omnipresent at or near  
655 the surface of the widespread remnants of the Stuart Range tableland and available in  
656 numerous outcrops and extensive lag gravels ('gibber plains', 'stony deserts'). The facies vary  
657 according to the parent material affected by the silicification (coarse- or fine-grained;  
658 abundant matrix material, or not, between the residual detrital grains). Illuviation structures  
659 enriched in titania are symptomatic of these facies. To find any specific characteristics of a  
660 particular facies in a particular location would seem to be difficult. On the other hand,  
661 groundwater silcrettes (quartzite, porcellanite and jasper of variegated colour) are available in  
662 outcrops and lag gravels along the creeklines that leading down to Lake Eyre. They have  
663 more distinctive characters and a detailed inventory may provide tracers of specific materials.

664         About 700km north of Coober Pedy and the Stuart Range, in the Todd River Plain  
665 near Alice Springs in central Australia, similar paleogeographic relationships occur. Here, in  
666 the scarp foot zone of a formerly extensive early Cenozoic pediment developed over the  
667 Amadeus Basin, mesas and buttes capped by the massive and impressive columnar facies of a  
668 pedogenic silcrete overlie a bleached and weathered regolith containing groundwater silcrettes  
669 (Milnes and Thiry, 1992; Milnes and Twidale, 1983). In distal parts of the pediment there  
670 are plateau remnants armoured by thick groundwater silcrettes and adjacent areas of silicified  
671 limestones (Fig. 17).

672         Here, also, the silicified facies are extremely varied and the associated bleached  
673 profiles are rich in white kaolinic clays and Fe oxides of variegated colours. Small mesas of  
674 fine quartzitic sandstone have been intensively quarried for tool-making. The steep scree-  
675 slopes around the hills and parts of the plateau surface are in places buried by thick deposits

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676 of discarded flakes and chips. The clear evidence of significant (and possibly long-term)  
677 exploitation provides the basis for characterising the source silcrete in detail and exploring the  
678 distribution of stone tools made from it.

679           However, the vast region, maybe 1 million km<sup>2</sup>, stretching from central South  
680 Australia to the central parts of the Northern Territory, essentially marking the western part of  
681 the Lake Eyre Basin (see Alley, 1998; Fujioka *et al.*, 2005) has innumerable outcrops and  
682 derivative lag gravels of pedogenic and various groundwater silcrettes, all of which could be  
683 exploited by Aboriginal people. Thus, there would be a significant challenge in progressively  
684 building an inventory of these materials sufficient to assist archaeological investigations in  
685 attributing artefacts to sources.

686           The nature and abundance of source material for tool-making has been noted from the  
687 first archaeological works in inland Australia by Aiston (1928), who stated “.. *it was so easy*  
688 *to make a tool that directly one failed to work satisfactorily it was discarded and a fresh tool*  
689 *made, a supply of stone material always being kept handy for this purpose...*”. The experience  
690 of Australian archaeologists comes both from stone tools excavated from archaeological sites  
691 and from ethnographic records of the experience of old Aboriginal people. Both types of  
692 study concluded that Aboriginal people were highly opportunistic in their use of source stone  
693 and took mostly varieties of local raw materials, including chert, chalcedony, jasper, silcrete  
694 (‘grey billy’ in the vernacular), quartzite, basalt, silicified wood as well as other igneous and  
695 metamorphic rocks (Cane, 1992; Flenniken and White, 1985). Nevertheless, small but  
696 significant amounts of non-local materials have been found dispersed over long distances and  
697 are thought to relate to migrations of prehistoric Aboriginal people during periods of drought  
698 (Gould and Sagers, 1985; Holdaway and Fanning, 2014; Mulvaney and Kamminga, 1999).

## 699           5.2 Paris Basin

700           The Paris Basin is bounded by Jurassic marine limestone overlain by thick chalk  
701 deposits with alternating continental and marine Tertiary deposits in the centre. The most  
702 remarkable feature of the Tertiary sequence is the interbedding of sandstone formations with  
703 limestones and marls. The present-day morphology of the Paris Basin, namely superimposed

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704 limestone plateaux, results from major uplift during the Pliocene and Quaternary which  
705 initiated downcutting and erosion of the Tertiary formations (Fig. 18). Silicified materials  
706 occur in almost all formations.

707           Pedogenic silcretes extend from the peripheral basement in the Central Massif and  
708 Brittany to the centre of the basin (Fig. 18). They developed along a paleosurface shaped by  
709 clastic discharge during the lower Eocene (Blanc-Valleron and Thiry, 1997). Silicification  
710 affects palaeoweathering profiles of clay-with-flint above Jurassic limestones and Cretaceous  
711 chalks, old alluvium to form thick puddingstones, and sandy clays that become the  
712 predominant silicified material at the border of the basin. The thickest silicified profiles (more  
713 than 15m thick) are found in grabens and channels that formed the lowlands (Thiry and  
714 Simon-Coinçon, 1996).

715           Groundwater silcretes developed in almost every Tertiary formation in the centre of  
716 the basin. Tightly cemented quartzite lenses occur in all sandy formations and silicified zones  
717 are present in every lacustrine and marine limestone formation (Thiry, 1999). Even the  
718 weathered materials topping the limestone plateaux contain large silicified features.  
719 Groundwater silcretes also developed in sandy and conglomeratic regolith materials topping  
720 the Mesozoic sequences bordering the basin. Recent groundwater cementation has been  
721 superimposed on former pedogenic silcrete profiles on the outer borders of the basin. It is  
722 likely that 5 to 10% of the outcropping formations in the basin are silicified, and this  
723 represents a considerable mass of easily accessible silcrete for potential exploitation.

724           Added to this inventory as well are Jurassic cherts and Cretaceous flints that occur all  
725 over the Paris Basin. They are *in situ* in the Mesozoic formations or occur in extensive areas  
726 covered by paleoweathering profiles of clay-with-flint and reworked into thick alluvial  
727 deposits within valleys that dissect the plateaux. These Jurassic cherts and Cretaceous flints  
728 generally account for the main component of lithic source material. Their identification was  
729 essentially based on micropaleontological determinations and was consequently limited in  
730 terms of pointing to source areas (Masson, 1981; Valensi, 1953).

731           Due to the predominance of cherts and flints, less interest has been applied to the  
732 recognition of various silcrete facies. Nevertheless, some investigations, in particular in the

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733 Paris Basin, identified several kinds of silcretes used for making tools. Fine-grained  
734 quartzitic silcretes from Fontainebleau have been recognized in flakes (Robin, 1974), used for  
735 polished axes (Bostyn *et al.*, 2012), and even to made into thin leaf-shaped Solutrean points  
736 (Sacchi *et al.*, 1996). These silcretes correspond to the uppermost facies, directly below the  
737 limestone cover, and are of glassy appearance due to the presence of microquartz together  
738 with quartz overgrowths forming the cement. Bartonian silicified lacustrine limestone has  
739 also been recognized (by means of its fossil content) in polished axes that are thought to have  
740 been dispersed up to 250 km (Bostyn *et al.*, 2012). Elsewhere, similar Bartonian silicified  
741 lacustrine limestone has been found together with artefacts made from meulière (weathered  
742 silicified limestone), silicified Lutetian marine limestone and quartzitic silcretes (Augereau,  
743 2008; Lanchon *et al.*, 2008; Mauger, 1985; Surmely, 2009).

744         Recent studies in and around the margins of the Massif Central, taking into account  
745 the petrography of cherts, flints and silcretes, including changes in response to their colluvial  
746 and alluvial reworking, made it possible to refine source areas (Fernandes, 2012; Surmely *et*  
747 *al.*, 2008). It has to be stressed that it is in basement areas (Massif Central and Bretagne)  
748 where the siliceous materials are scarce that the sourcing studies were the more skilful and the  
749 most successful. Detailed studies of flint and chert cortices from the Massif Central made it  
750 possible to consider natural dispersion of the primary materials and consequently to determine  
751 more precisely the source areas (Fernandes, 2012). Silcrete deposits exist also in these  
752 basement areas, although scattered and variable in facies. Detailed studies have been able to  
753 track the dissemination of these materials within the massif as well as those imported from the  
754 peripheral basins (Aubry, 1991; Célérier, 1990; Dabard *et al.*, 2012; Delvigne, 2012; Surmely  
755 *et al.*, 2008; Wragg-Sykes, 2014).

## 756 **6 Identifying provenance**

757         There are certainly specific macroscopic criteria that could be used to help identify  
758 sources of silcrete tools, but most of the time these investigations are ambiguous. Colour,  
759 grain size and distribution, and the character and aspects of the break, are generally not  
760 specific. The morphologies of silcretes are often typical at the outcrop scale, but not at the  
761 artefact scale.

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762           Micromorphological and petrographical features of silcretes are more reliable criteria  
763 of origin and potential provenance. However, identification of provenance from these  
764 features requires a comprehensive inventory of actual local and regional source materials, a  
765 sound geological knowledge of these occurrences, and an accompanying database of  
766 micromorphological and petrographical observations of representative samples. This is  
767 conceivable on a local scale but is much more difficult to manage on the regional scale of  
768 societal exchanges. Geochemical criteria, particularly the concentrations of indicator trace  
769 elements, may be helpful. Dutkiewicz *et al.* (2015) have analysed major and (particularly)  
770 trace elements in both opal and host rocks in Australia in order to identify the possible origin  
771 of the silica. This approach could potentially be extended to silcretes although, in pedogenic  
772 silcretes, the complex of silica forms and associations with microcrystalline anatase and other  
773 phases, and the multiplicity of silcrete facies, even on the scale of an outcrop, would pose a  
774 significant challenge. Moreover, Holdaway and Fanning (2014) and others have pointed out  
775 the complexities presented by the suite of stone artefacts in archaeological sites wherein there  
776 are local and foreign sourced materials representing an interplay of unknowable activities.

777           Finally, questions of the extent of post-discard alteration and degradation of  
778 archaeological materials has to be addressed. Exoscopic studies of the surfaces of stone tools,  
779 which are often altered in various ways by patina evolution, can provide few critical data  
780 (Fernandes, 2012; Thiry *et al.*, 2014).

781           The difficulty for archaeological studies is to precisely characterize the many varieties  
782 of silica-rich source rocks for stone tools. In the past, only stratigraphic characters and fossils  
783 have been used to determine the provenance of tool materials (Mauger, 1984). This was of  
784 limited use because the stratigraphic levels are relatively widespread. More detailed, multi-  
785 criteria identification is necessary and essential for understanding trade and acquisition  
786 strategies involving stone tools.

787           Progress is possible by taking into account precise micromorphological characteristics  
788 that will potentially point to source sites. However, this approach requires an insightful  
789 inventory and petrographic data led by archaeological studies (Fernandes, 2012). Inventories  
790 of this type are yet not available. Even existing geologic and geomorphological maps do not

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791 make reference to the specific characters of silcretes, silcrete profiles, or micromorphological  
792 features of particular facies, that could be key to these types of investigations. The  
793 foundations of such inventories exist, but a new generation of linked archaeological and  
794 geological research would be a next step.

## 795 **7 Summary**

796 Several forms of silcrete are recognised in remnants of paleolandscapes in Europe,  
797 Africa and Australia dating from the Mesozoic to the present day. Their chemical,  
798 mineralogical and physical (microstructural) composition accounts for their persistence and  
799 for the major influence of some silcretes in defining the unique morphology of landforms. All  
800 silcretes, of which there are 4 - 6 main types, formed as a result of the introduction and  
801 precipitation of silica, for example via groundwaters, or the mobilisation and precipitation of  
802 silica from pre-existing minerals via alteration and the leaching of other elements, for  
803 example in acidic environments. Each is characterised by a variety of silica polymorphs,  
804 including quartz, various forms of microcrystalline and cryptocrystalline quartz, and various  
805 forms of opal (opal-CT, opal-A), in particular micromorphological arrangements. The main  
806 categories of silcrete originated in pedogenic or groundwater environments.

807 Other than their mode of occurrence and morphological appearance, it is the  
808 microstructural composition of silcretes, dictated by mineralogical and micromorphological  
809 features that derive from their origin, that provides the basis for understanding their physical  
810 and mechanical properties (which relates to the attractiveness of some forms to prehistoric  
811 peoples for the making of effective tools). However, it is well recognised that there may have  
812 been other practical and even aesthetic or symbolic dimensions to such choices.  
813 Investigations of the provenance of stone tools, other than in a local area, will be challenged  
814 by a need for regional inventories of silcrete facies.

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1116 **Figure captions**

1117

1118 Figure 1 - Rate of dissolution and asymptotic solubility of the different silica varieties at 25°C  
1119 (after Siffert 1967). Opal and chalcedony show variable solubility according to their  
1120 respective crystallinity.

1121 Figure 2 - Quartz and chalcedony solubility versus temperature (computed from geochemist's  
1122 workbench program; Bethke, 2002). Quartz solubility decreases to more than half of its  
1123 initial value between 12.5 and 0°C.

1124 Figure 3 – Solubility of amorphous silica in aqueous salt solutions at 25°C (after Marshall,  
1125 1980)

1126 Figure 4 – Alumina and silica solubility versus pH (computed from PHREEQC program, U.S.  
1127 Geological Survey; Parkhurst and Appelo, 1999). Silica is insoluble at acidic and neutral  
1128 pH that appear favourable for development of silcretes, whereas alumina stability is  
1129 restricted to neutral pH.

1130 Figure 5 – Eh/pH diagram for Fe-O-H system showing the reaction path leading to ferrolysis  
1131 after having introduced some oxygen into the Fe<sup>++</sup>- rich solution. Diagram at 25°C in  
1132 equilibrium with atmospheric O<sub>2</sub> and CO<sub>2</sub>, and [SO<sub>4</sub><sup>--</sup>]= 10<sup>-7</sup> (after Garrels and Christ,  
1133 1965).

1134 Figure 6 –Schematic geological section through the Beauce Plateau. Quartzitic silcrete pans  
1135 are limited to the outcrop zone of the Fontainebleau Sand. The size of the silcrete lenses  
1136 (black wedges) is exaggerated for illustration.

1137 Figure 7 – Schematic model of successive cycles of groundwater-related quartzitic silcrete  
1138 formation in the Fontainebleau Sand. The thickness and the slope of quartzite pans are  
1139 exaggerated for illustration.

1140 Figure 8 - Schematic diagram of the Calcaire de Champigny Formation (upper Eocene)  
1141 showing the distribution and shapes of silicified zones (Plateau of Brie, France). Note  
1142 similarity between the distribution and shapes of the silicified zones and dissolution  
1143 features of the limestone.

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1144 Figure 9 - Larkins Folly section exposed in bulldozer costeans in Coober Pedy opal field. The  
1145 bleached Cretaceous formations contain termite burrows and alunite nodules. There are  
1146 two superposed levels of groundwater silicification. In addition, at the top is a  
1147 pedogenic silcrete disrupted by vertical pipe-like structures, and an overlying laminar-  
1148 structured red-brown hardpan.

1149 Figure 10 – Sketch of the macromorphological organisation of a pedogenetic quartzose  
1150 silcrete and distribution of secondary silica and dissolution structures.

1151 Figure 11 – Thin sections of groundwater silcrettes (transmitted light, crossed polars). (A)  
1152 Fontainebleau quartzitic silcrete in which original detrital quartz grains are overgrown  
1153 by secondary silica, producing triple-point junctions and thus a compact mesh that  
1154 results in a rough fracture. (B) Stuart Creek quartzitic silcrete with chalcedonite deposits  
1155 in pore spaces produces a lustrous break on knapping.

1156 Figure 12 – Thin sections of silicified limestone (Paris Basin): two views (A) and (B):  
1157 transmitted light, crossed polars. The larger crystals are euhedral quartz that crystallized  
1158 in voids (v) by precipitation from water, the speckled areas are microcrystalline quartz  
1159 ( $\mu$ Q) replacing primary carbonate and the black areas are remnant carbonate (Ca). The  
1160 images show the nature of the silicified “front” (dotted line).

1161 Figure 13 – Thin sections of porcellanites (transmitted plane polarised light). (A) Stuart  
1162 Creek, South Australia. The dark granular matrix (Op1) is silicified silty claystone.  
1163 Voids (v) and channels contain successive silica deposits (Op2), firstly brown opal then  
1164 chalcedonite. The latter also impregnates the matrix and results in a compact material  
1165 with a lustrous fracture. (B) Sancerre, southern Paris Basin. The dark opal matrix (Op1)  
1166 has recrystallized into clear homogeneous microquartz ( $\mu$ Q); the opal deposits in the  
1167 voids (Op2) remain unaffected. This type of porcellanite provides sharp flakes, like  
1168 flints, on knapping.

1169 Fig 14 – Thin sections of pedogenic silcrettes, Paris Basin. (A) matrix of amoeboid quartz  
1170 microcrystals in net-like arrangement generates a hard and lustrous fracture on knapping  
1171 (transmitted light, crossed polars). (B) and (C) nodular facies with illuviation cutans (il)  
1172 recrystallized into microquartz plus titania microcrystals; nodules are rimmed by a

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1173 cortex enriched in titania (arrow). Channels remain empty (v) or are cemented with sub-  
1174 euhedral quartz crystals (Q). (Transmitted plane polarised light).

1175 Figure 15 - Schematic view of the Beda pediment showing the distribution of pedogenic  
1176 silcrete facies. Facies change completely at a 100 m scale (after Milnes and Thiry,  
1177 1992).

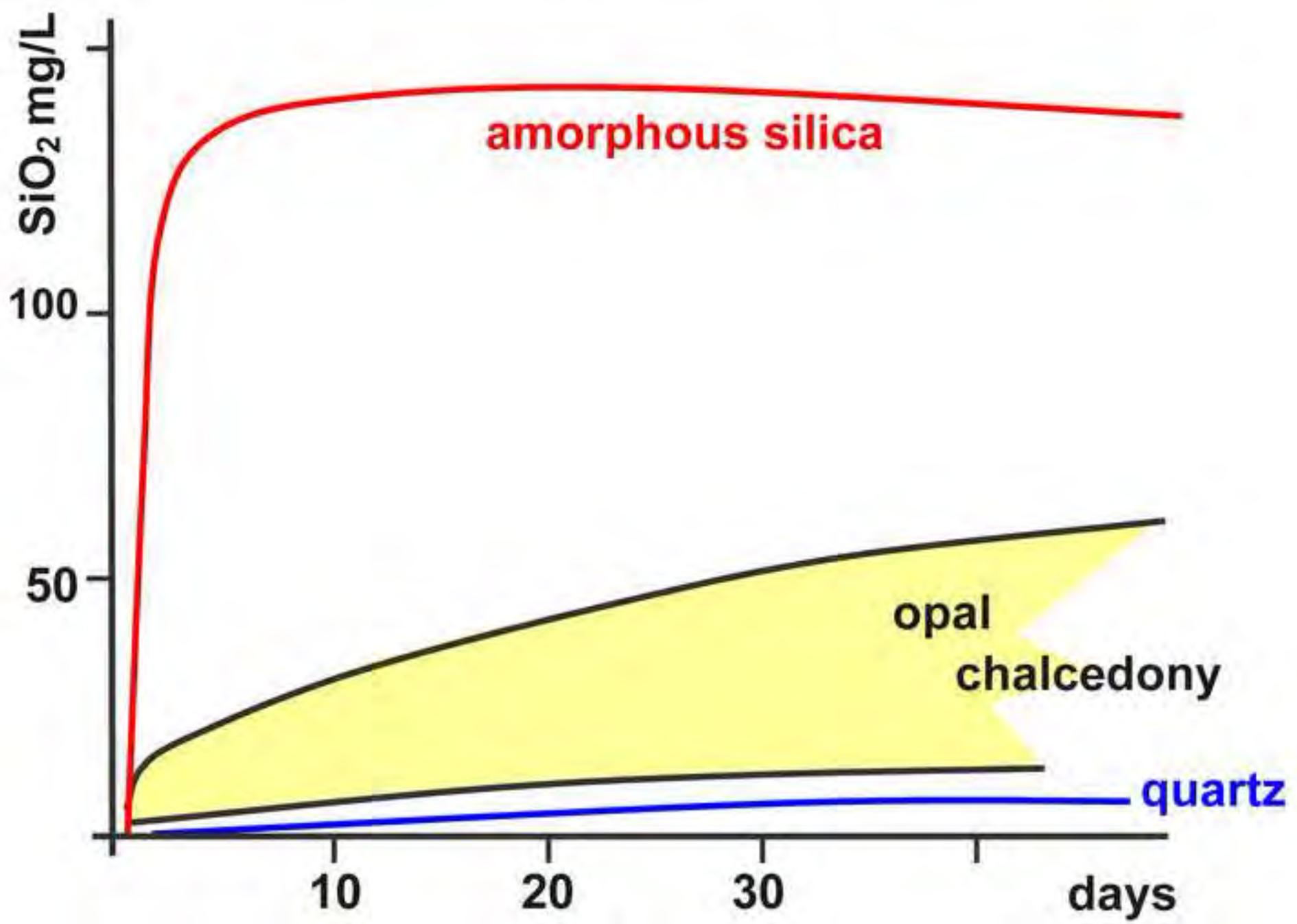
1178 Figure 16 - Schematic morphostratigraphic sketch of landscapes from the Davenport Range,  
1179 across the Stuart Range, to the Eucla Basin, South Australia, showing silicification in  
1180 relation to different weathering features and their distribution in relation to landsurfaces  
1181 and geology (after Simon-Coinçon *et al.*, 1996).

1182 Figure 17 –Schematic section across the Todd River Plain, southeast of Alice Springs, central  
1183 Australia, showing the location of different silcrete facies (after Milnes and Thiry,  
1184 1992).

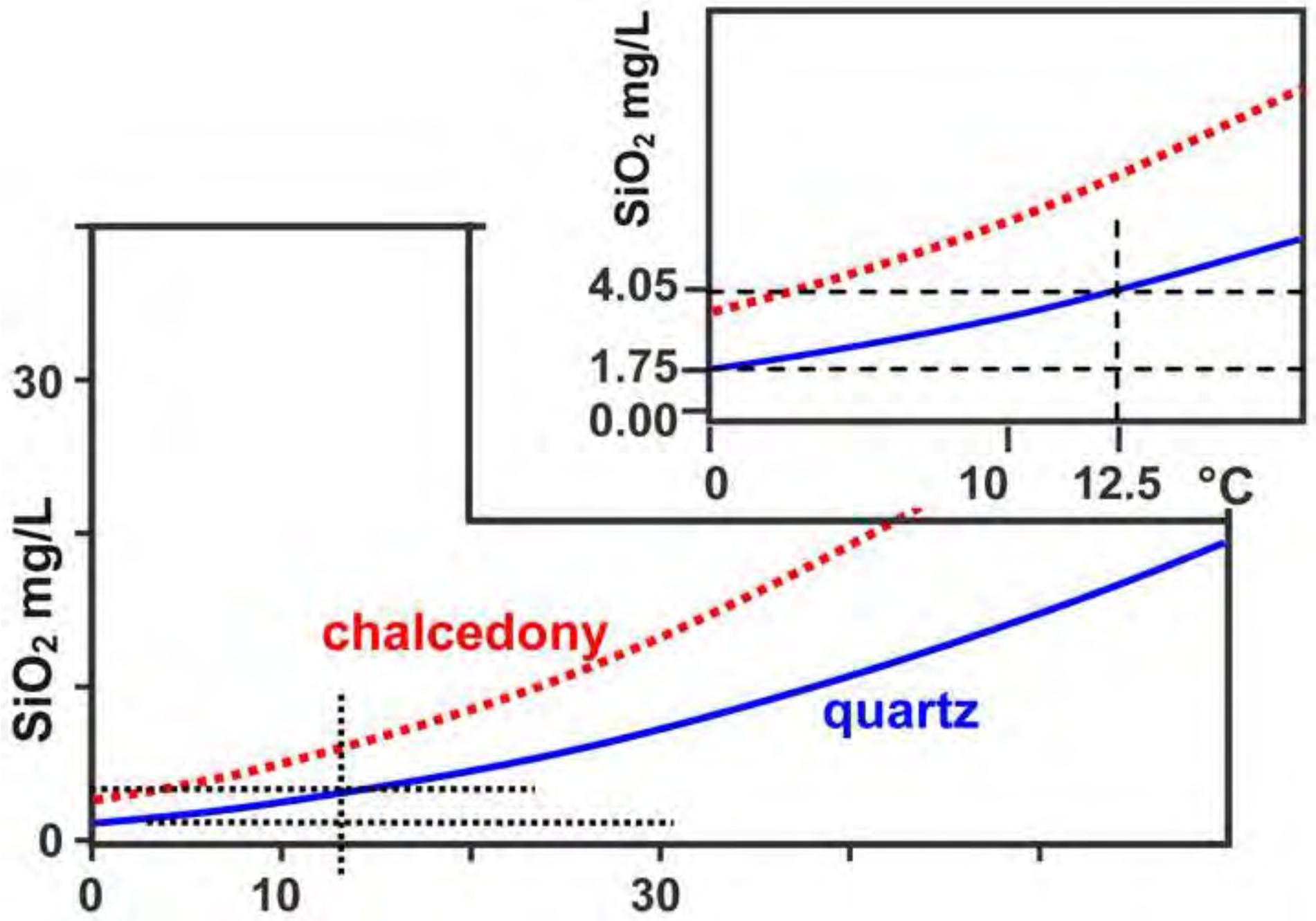
1185 Fig. 18 – Schematic geomorphological section across the Paris Basin, showing the  
1186 distribution and relationships of silcrete facies.

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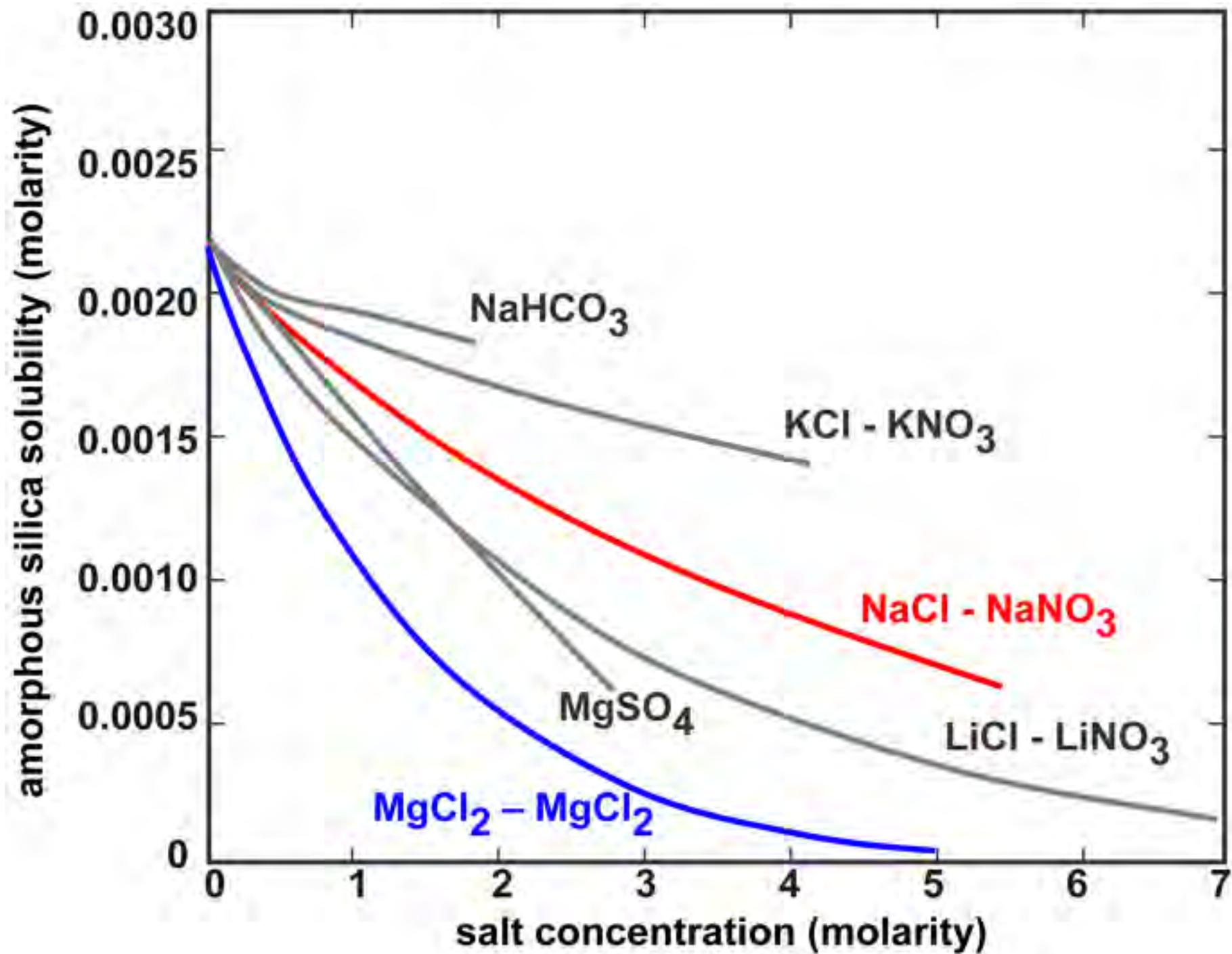
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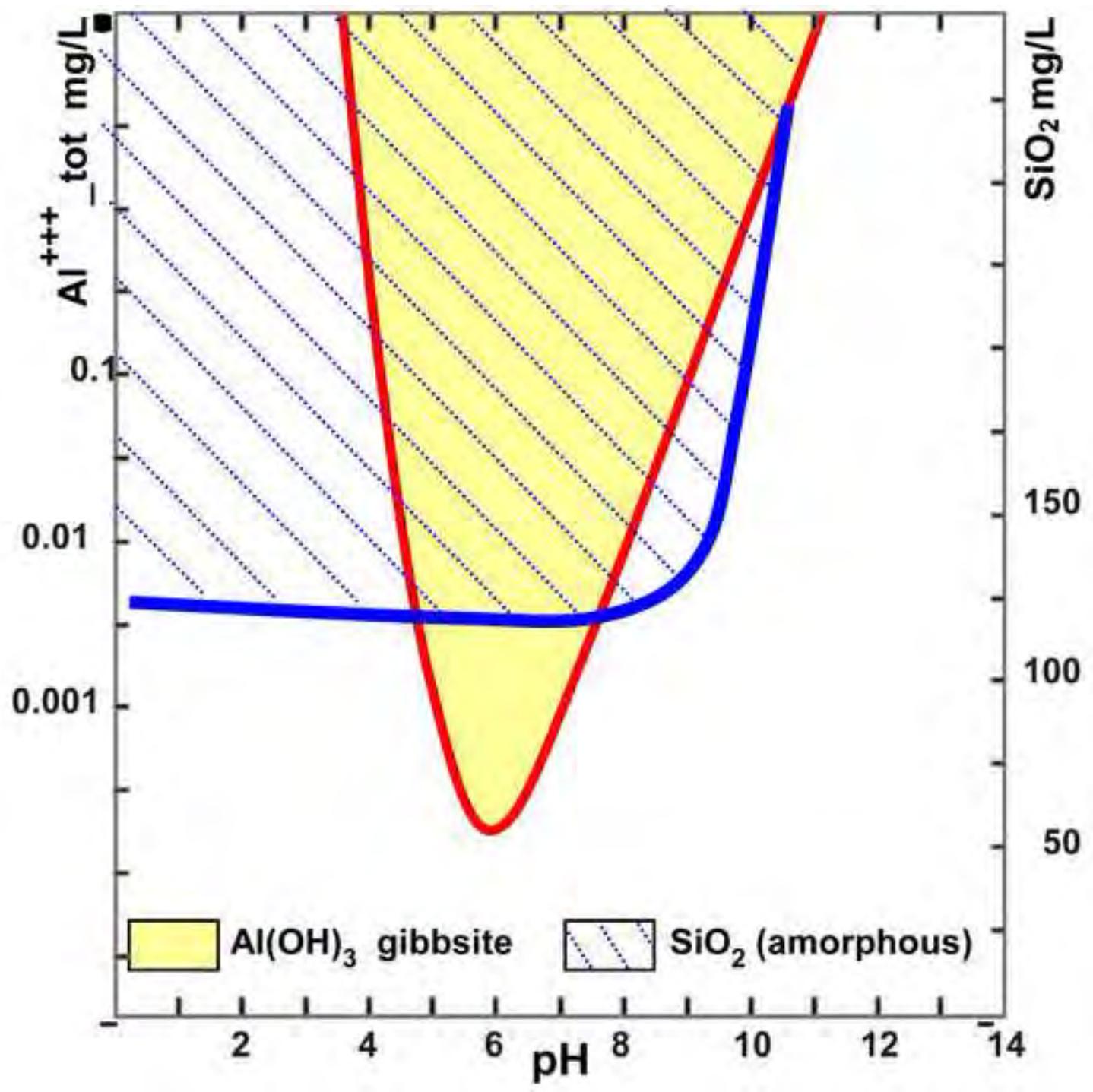


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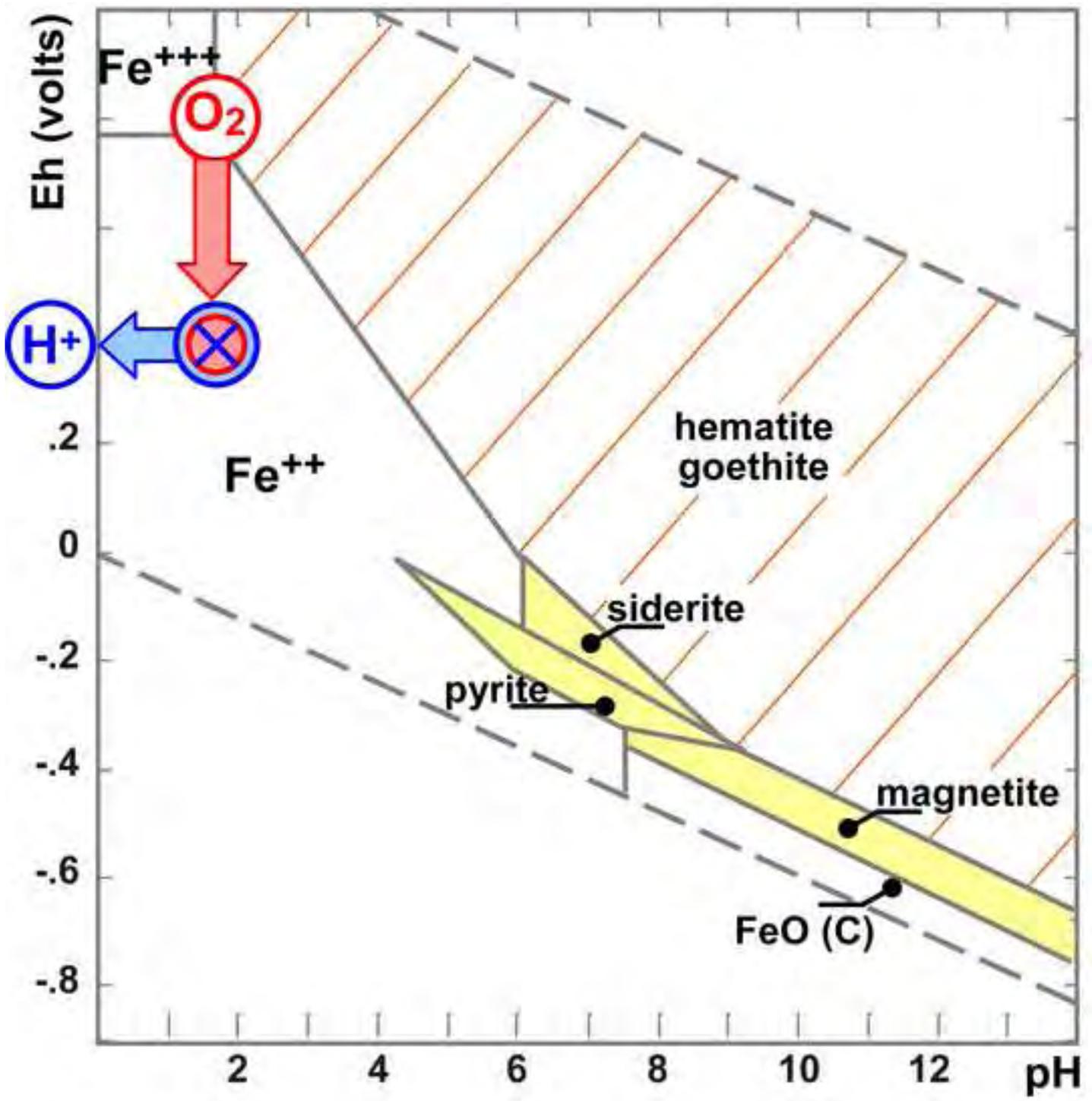
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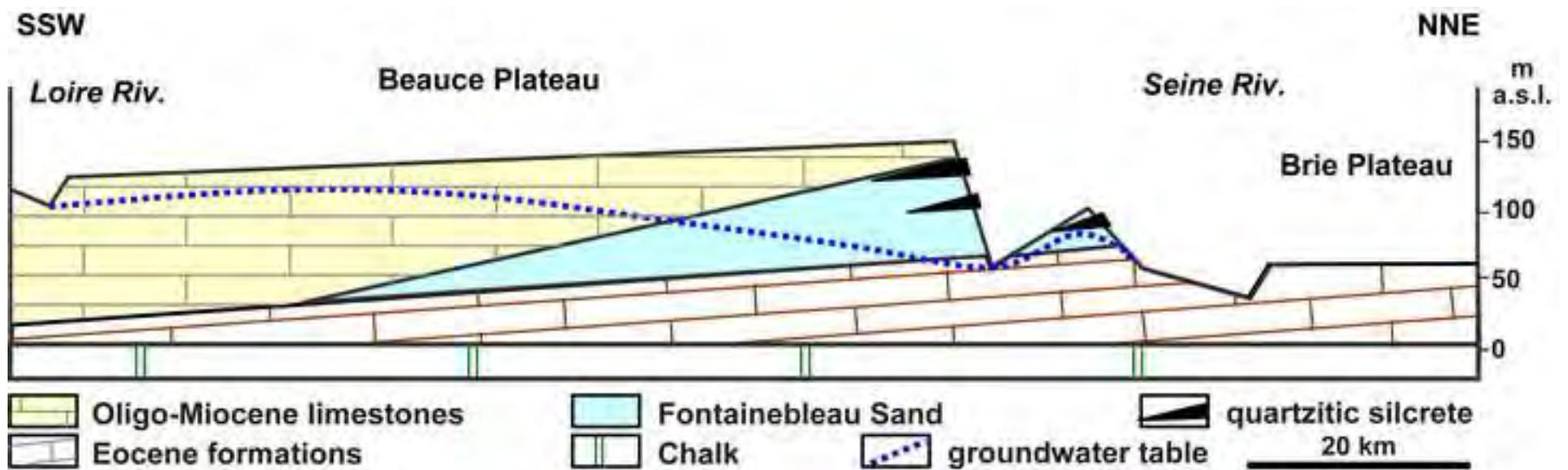


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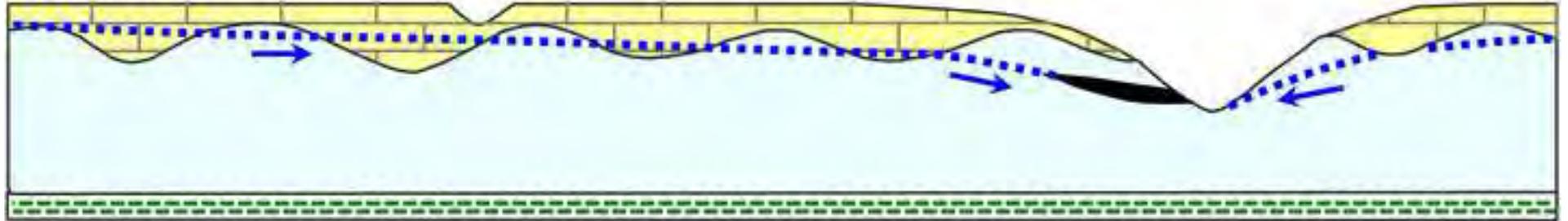


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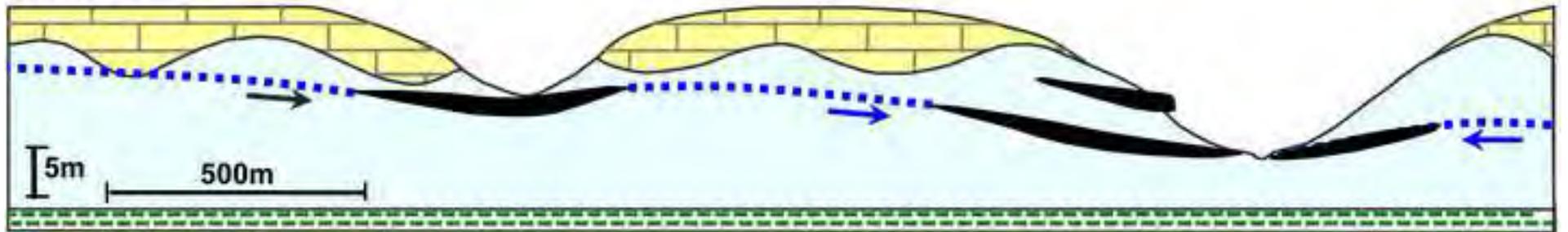


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### STAGE 1



### STAGE 2



marl



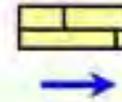
quartzitic silcrete



sand



water table



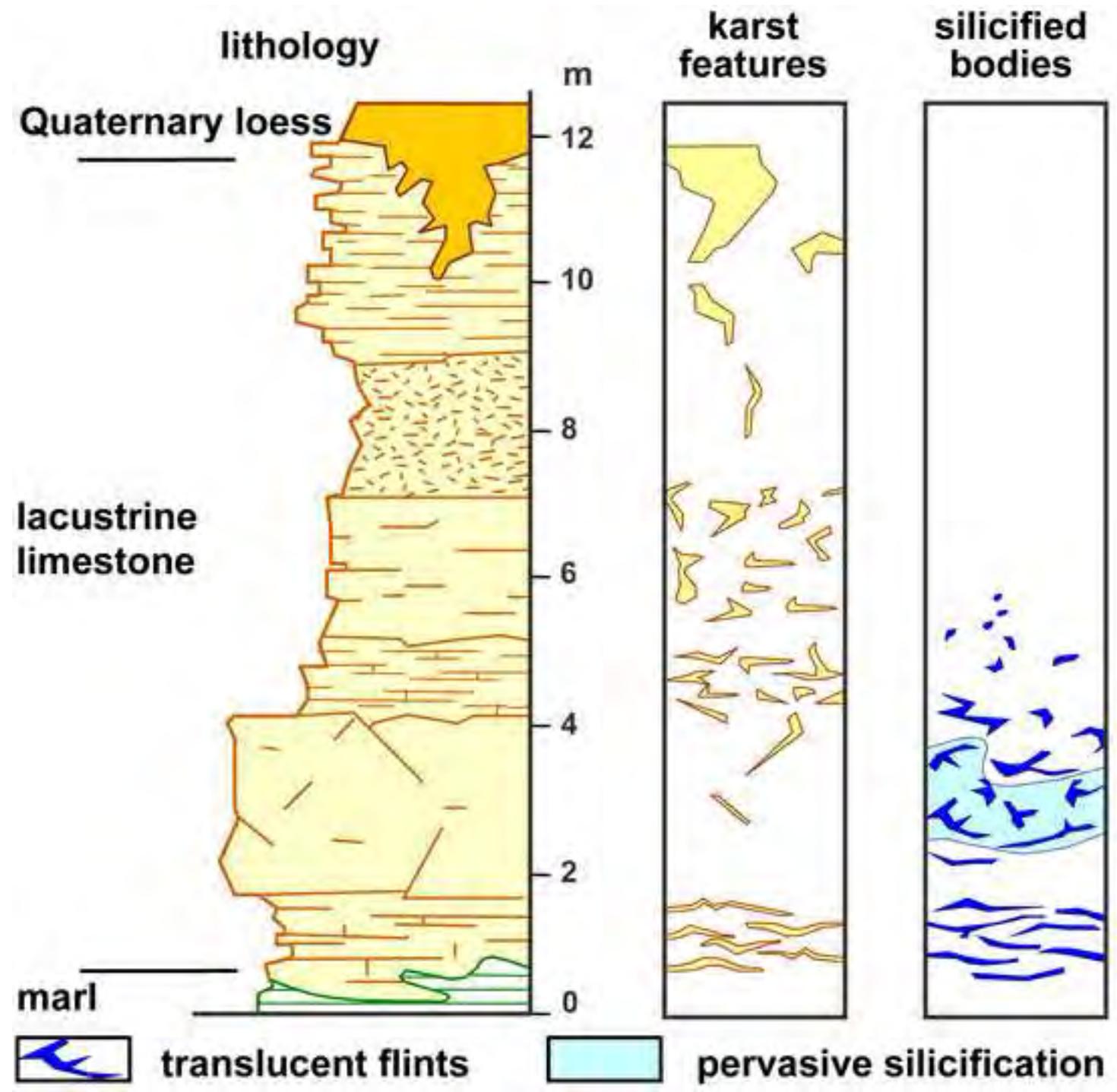
limestone

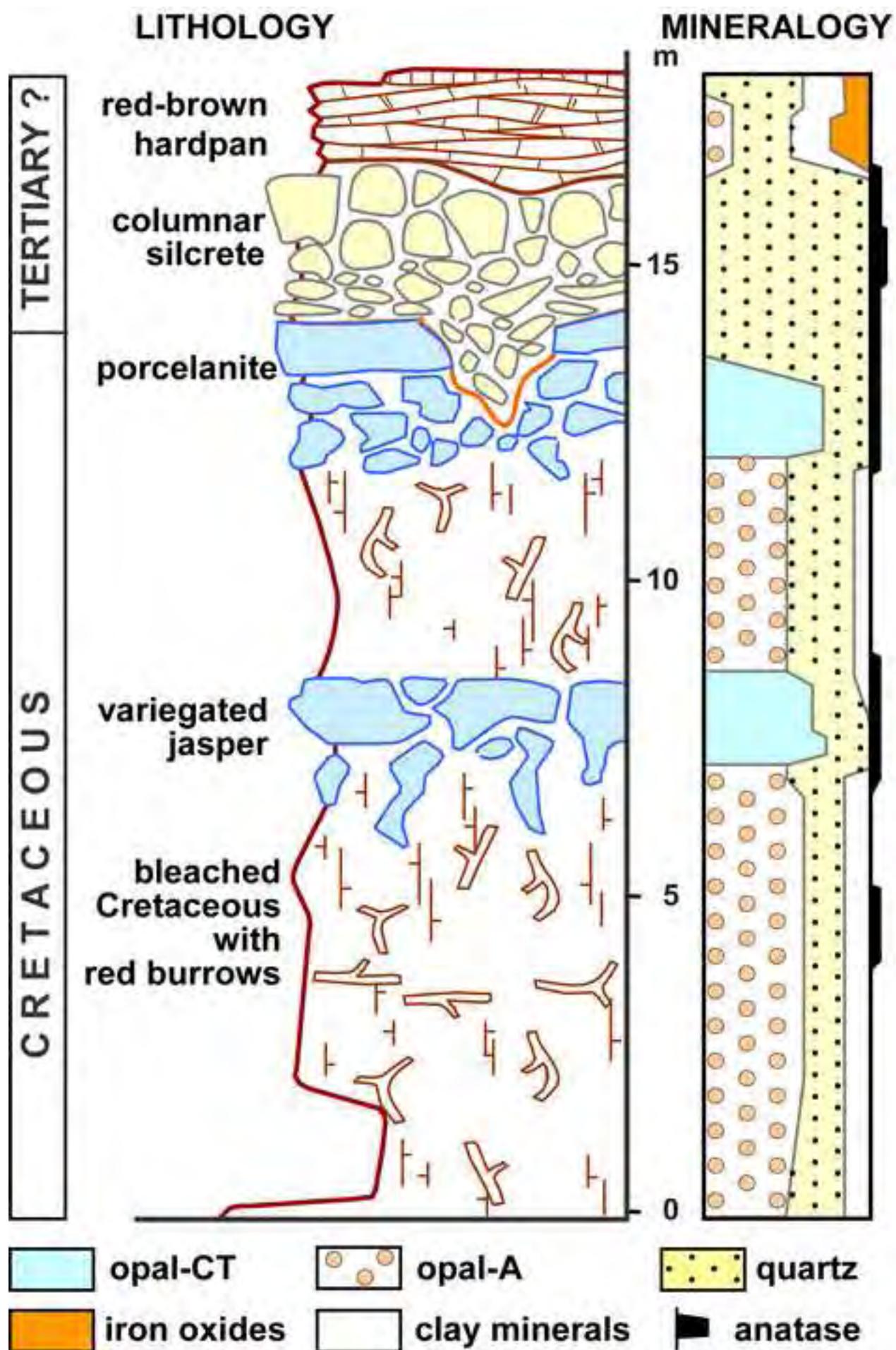


groundwater flow

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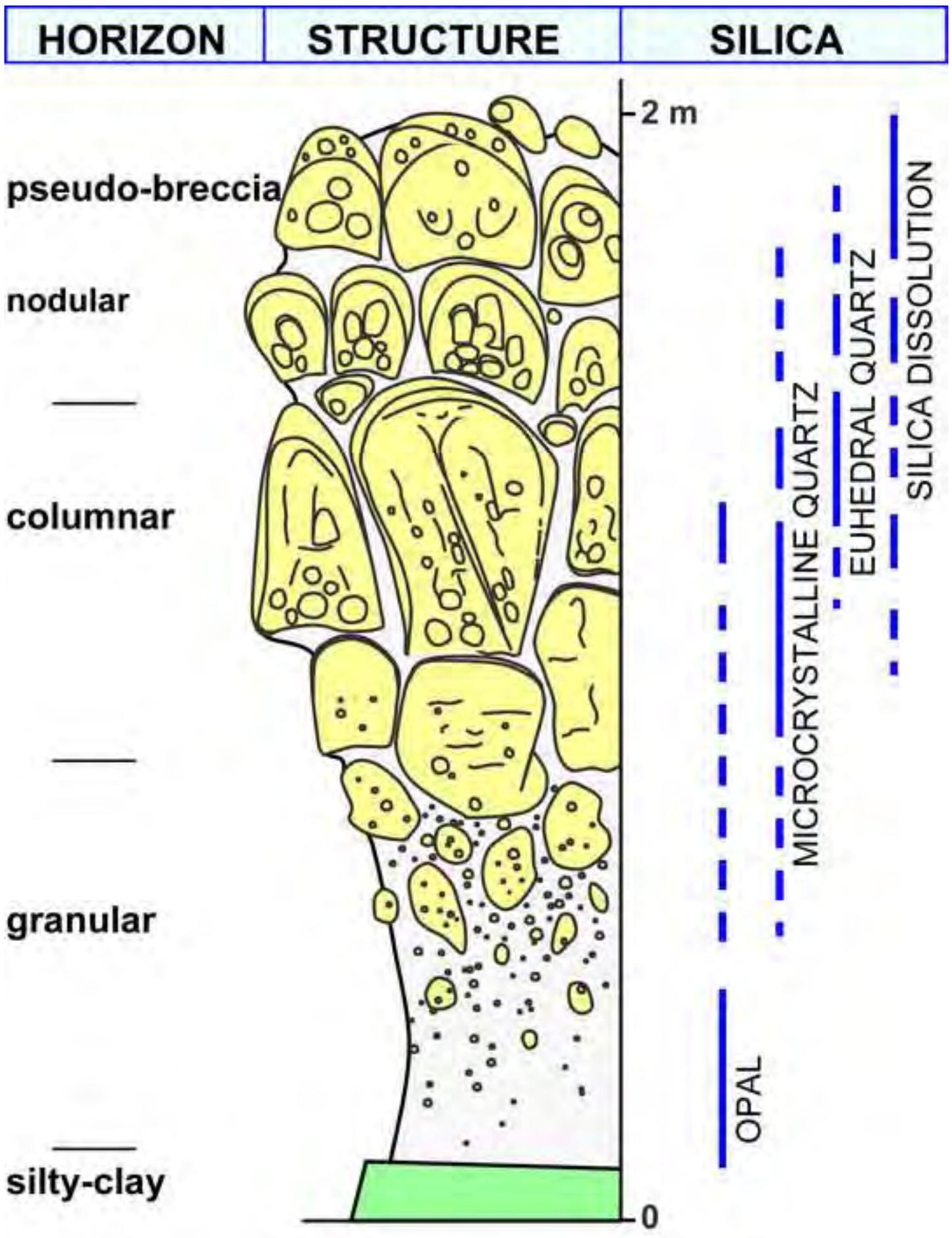
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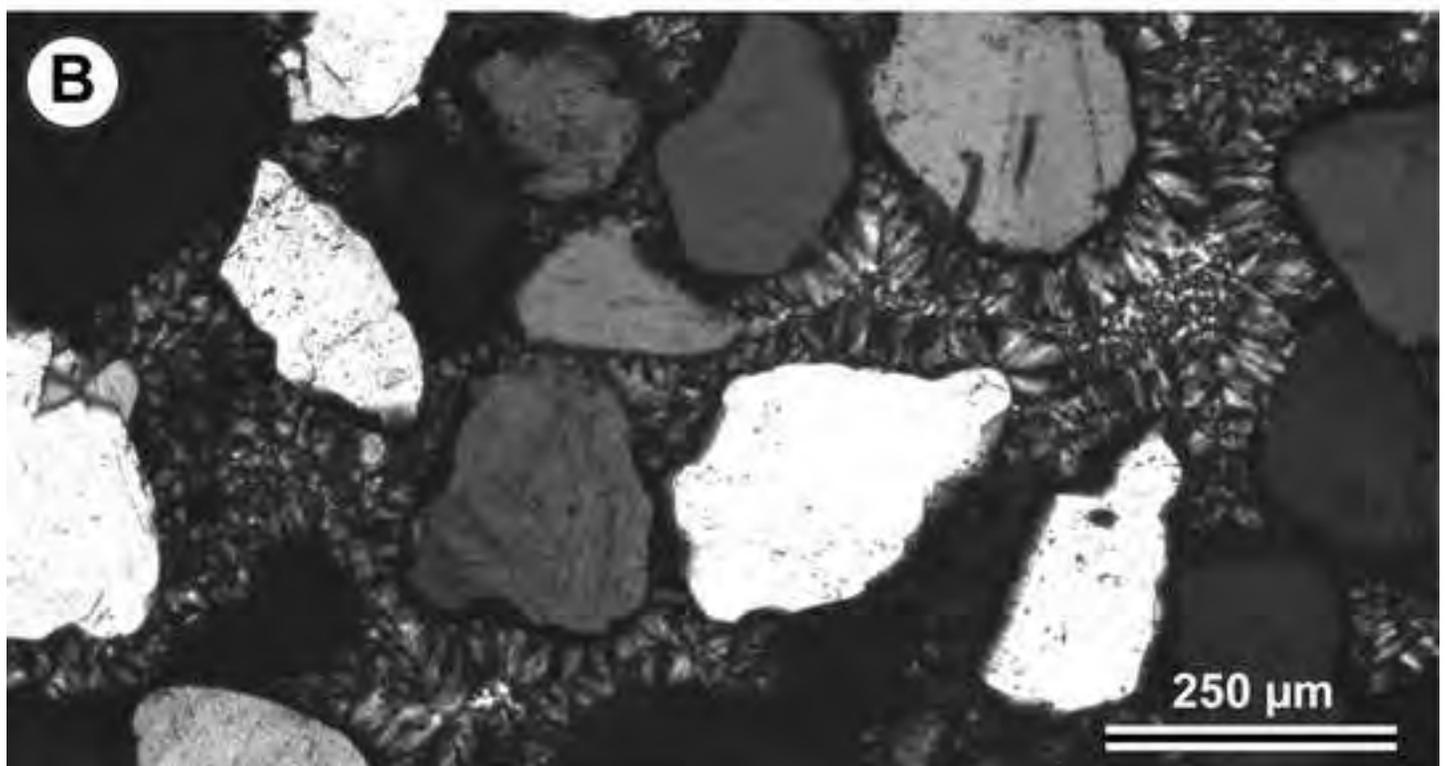
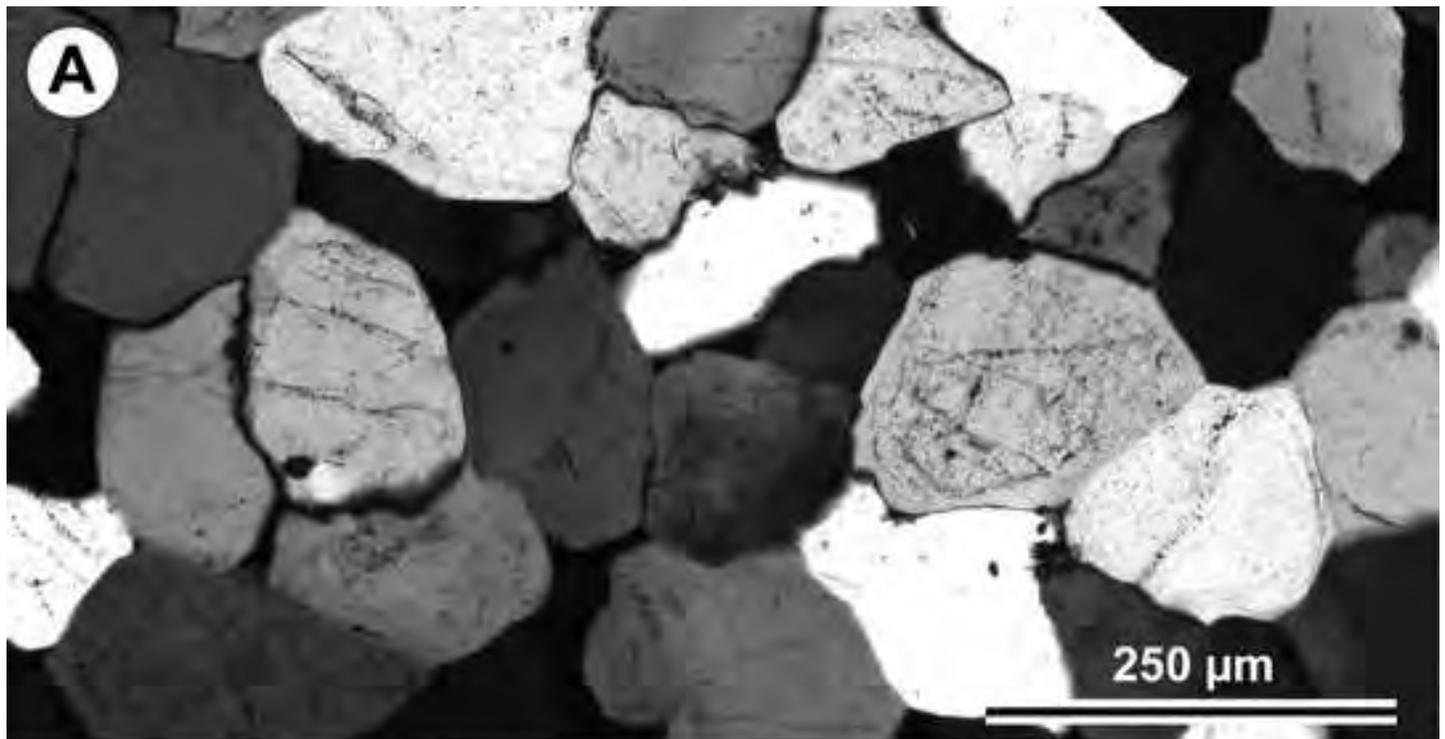


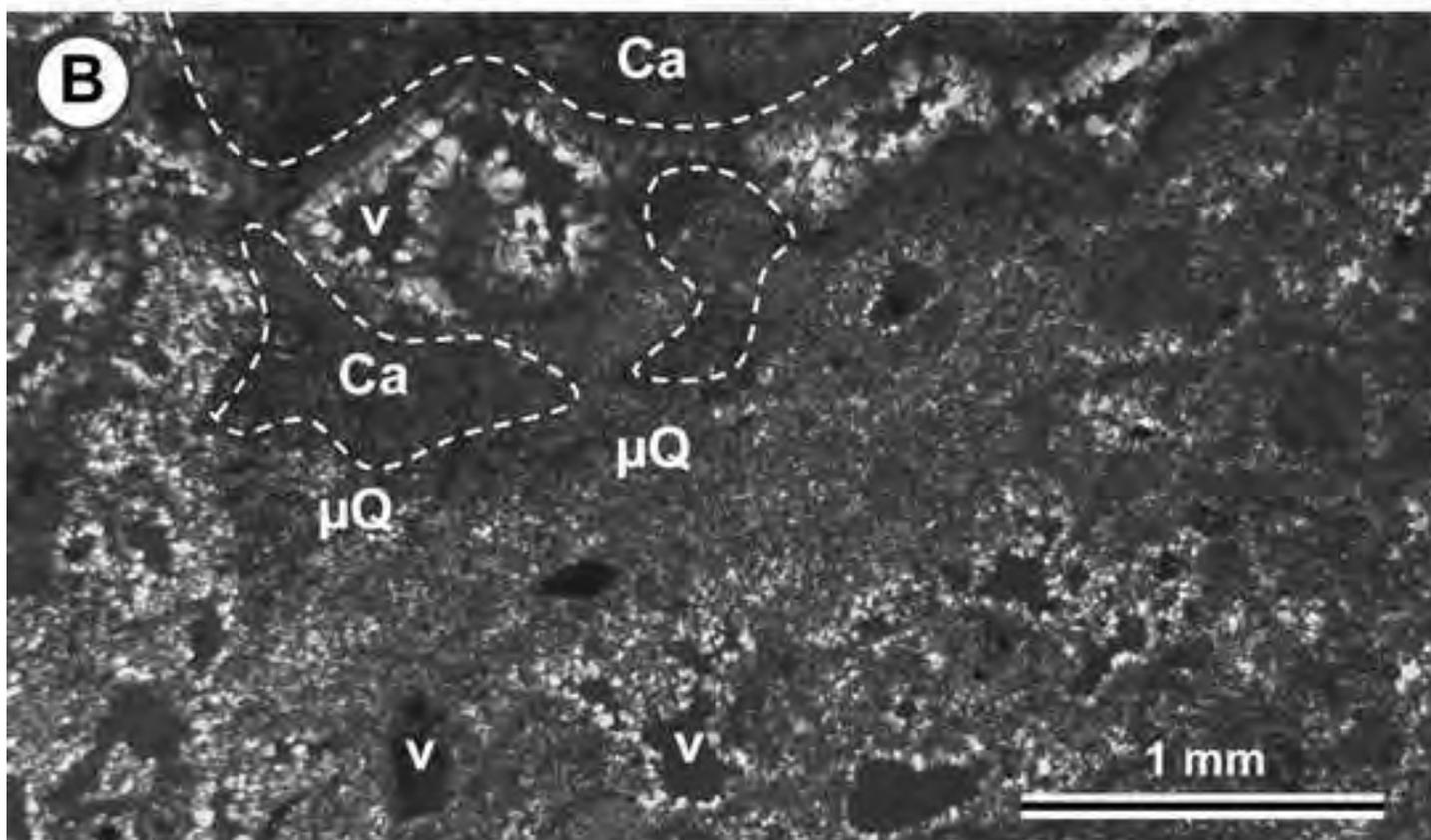
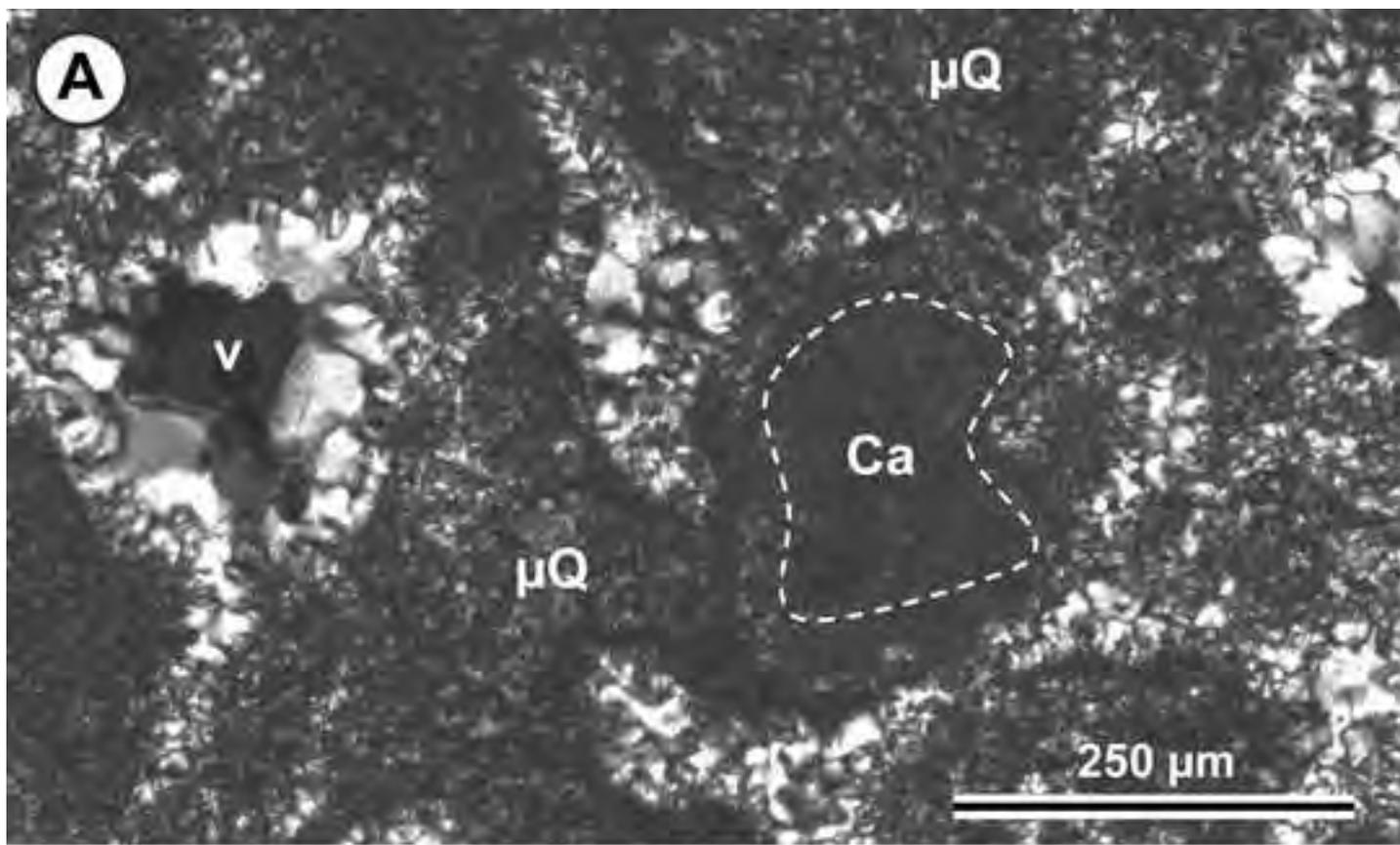


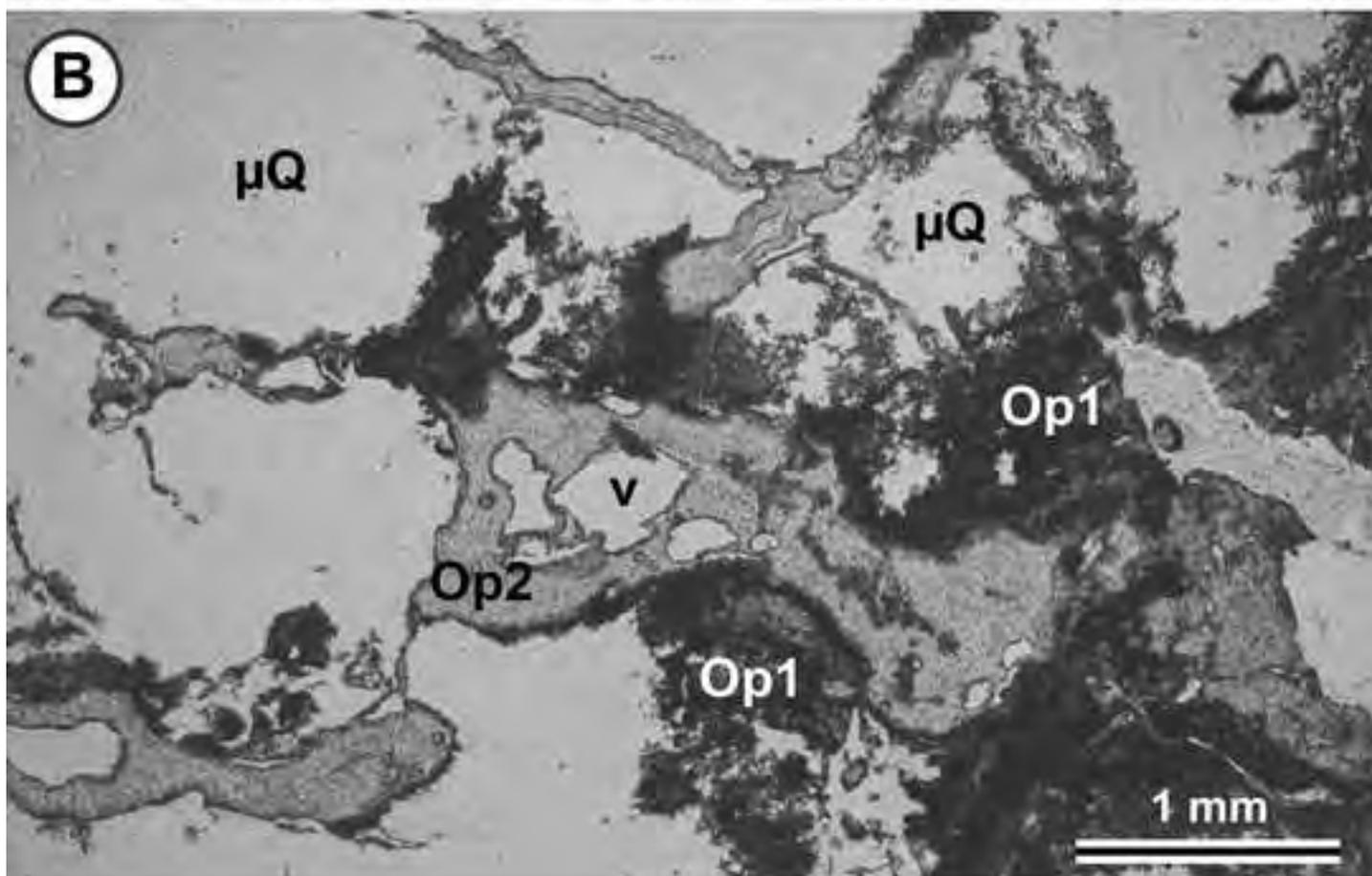
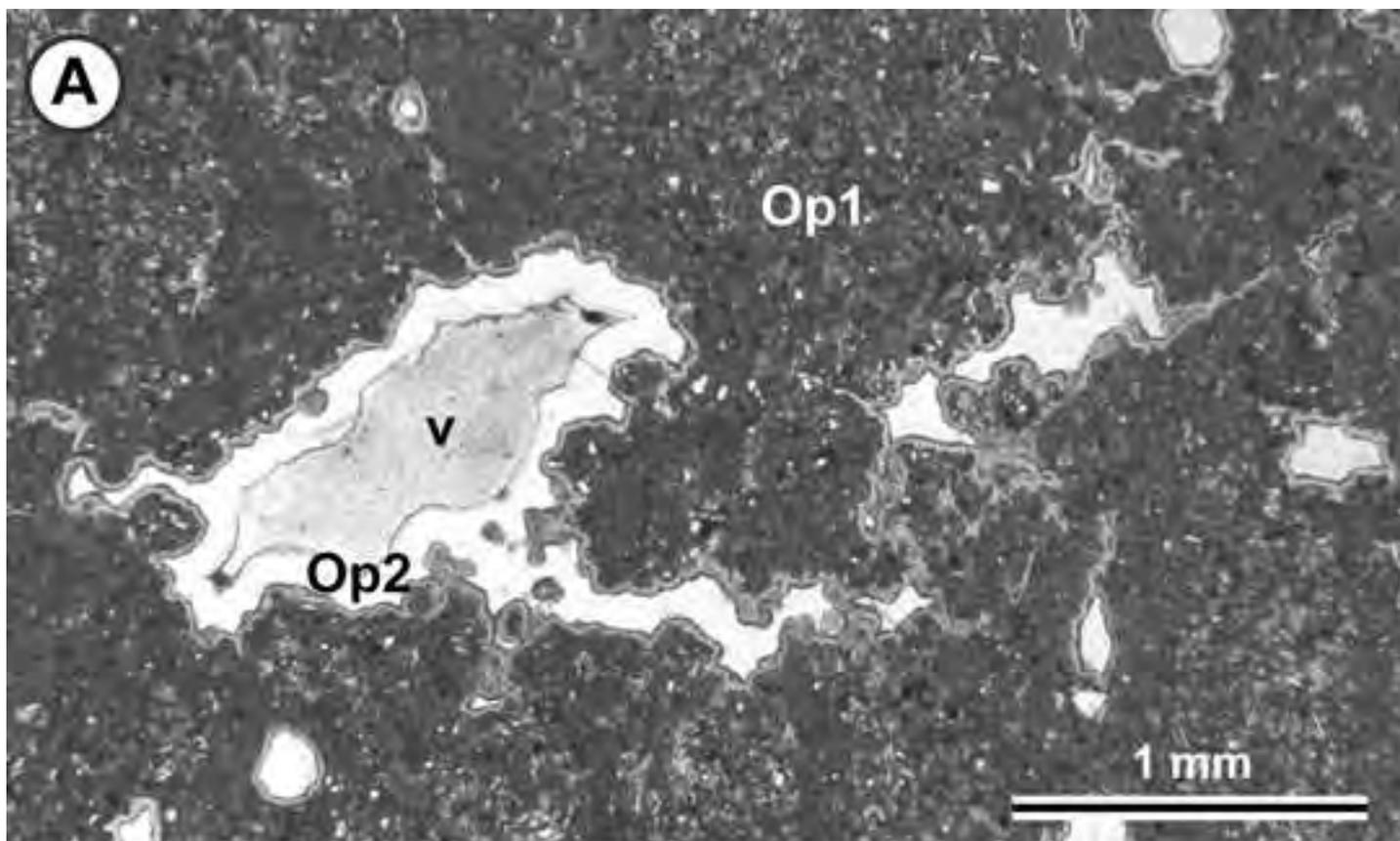
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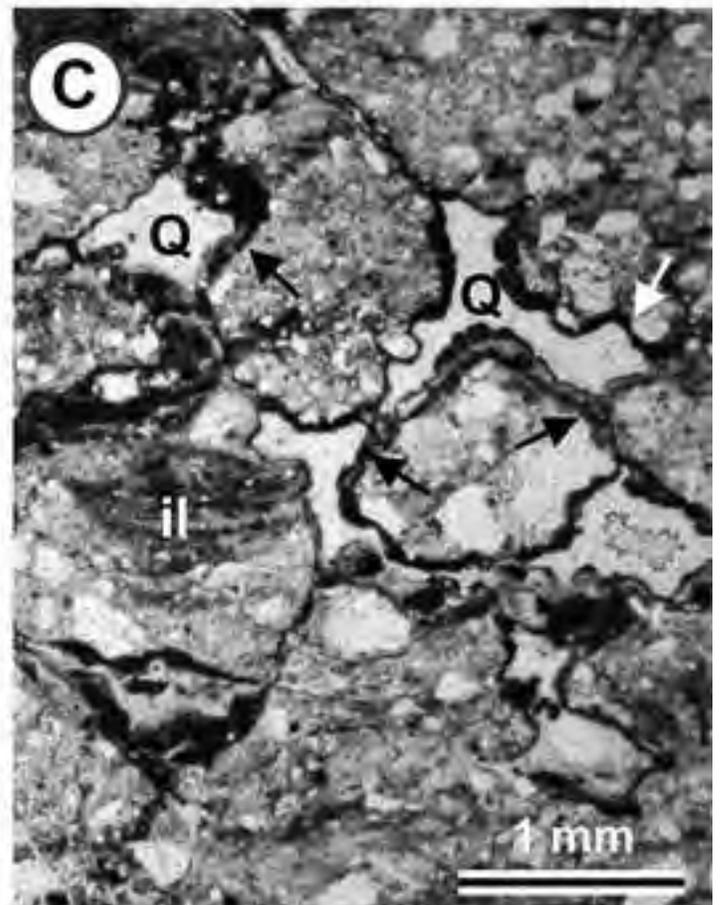
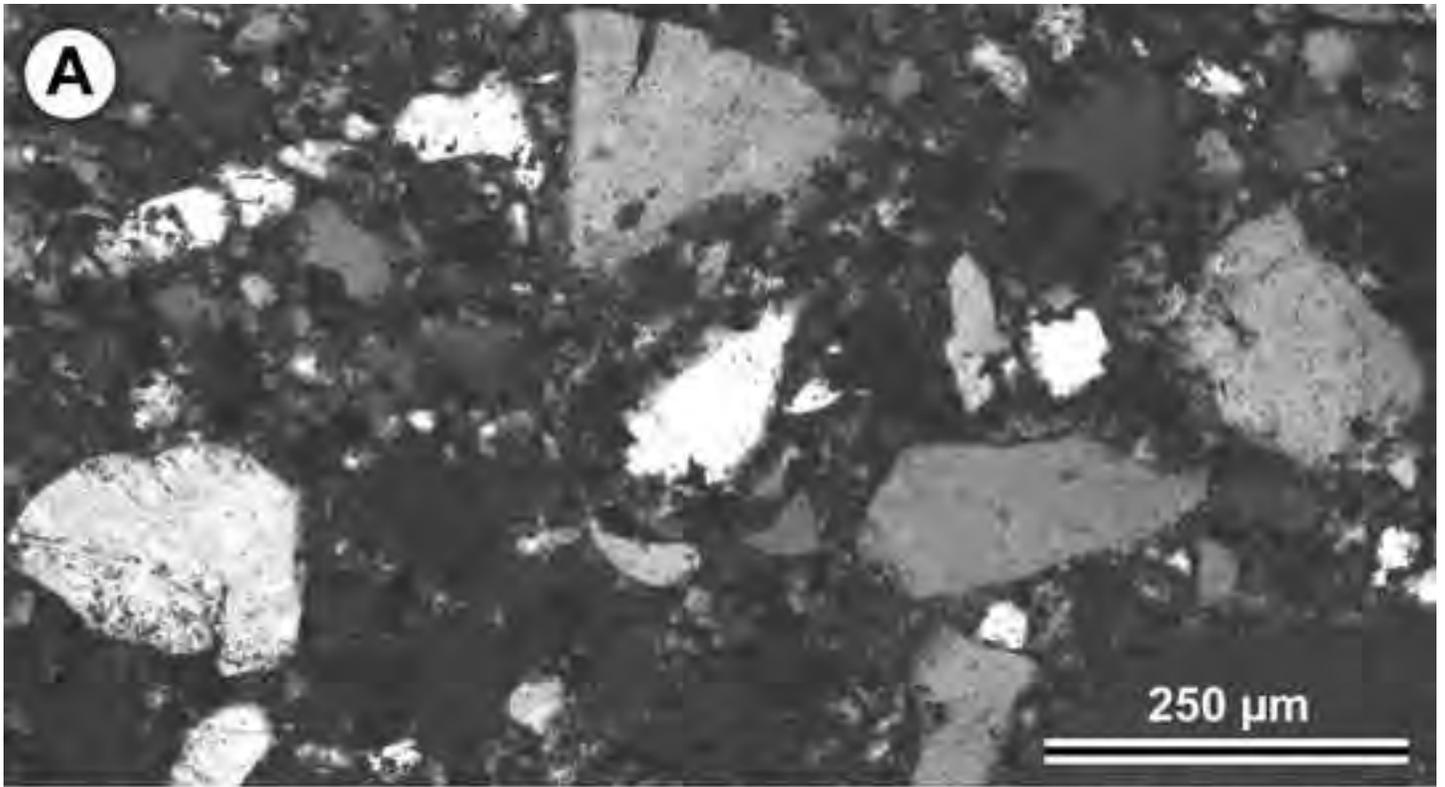
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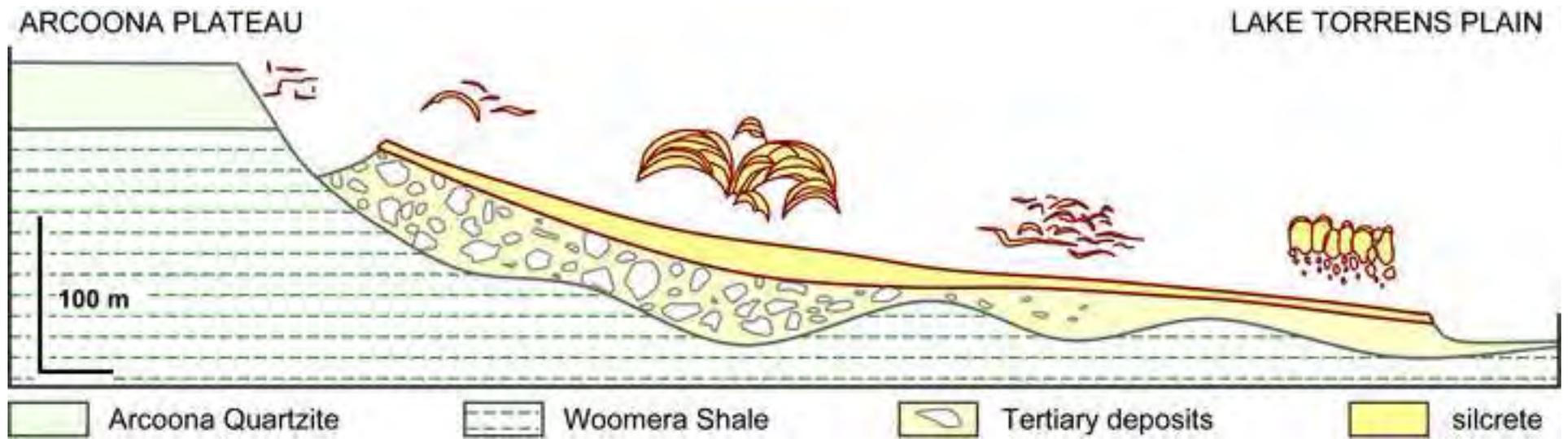




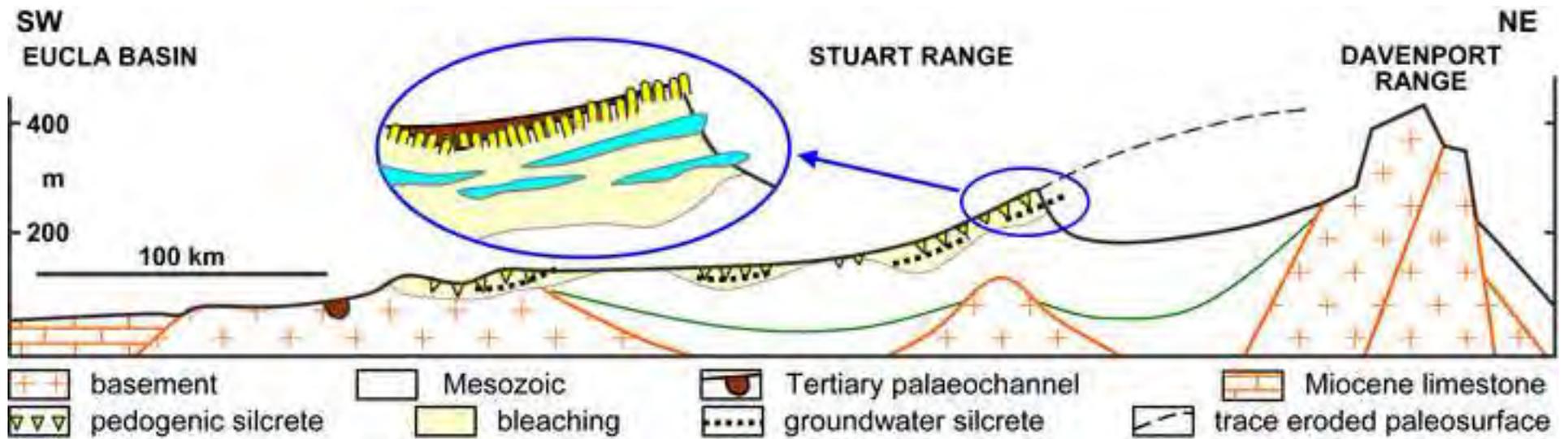




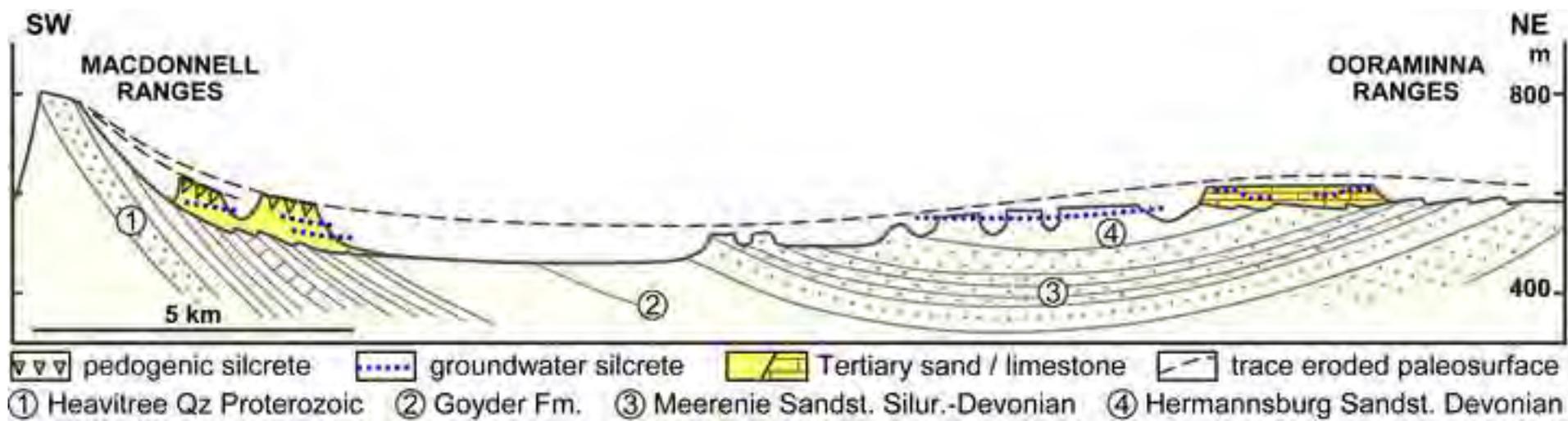
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