

Department of Applied Geology

**The Role of Biological and Non-biological Factors in the Formation
of Gold Anomalies in Calcrete.**

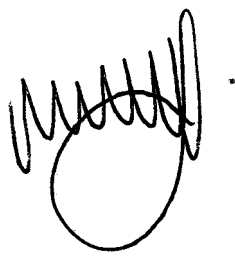
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**This thesis is presented for the Degree of
Doctor of Philosophy
of
Curtin University**

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Declaration

To the best of my knowledge and belief this thesis contains no material previously published by any other person except where due acknowledgment has been made. This thesis contains no material which has been accepted for the award of any other degree or diploma in any university.

A handwritten signature in black ink, consisting of several vertical, wavy lines that curve to the right, ending in a small dot.

Signature.....

Date.....8th December 2011.....

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I express gratitude to my co-authors of my submitted papers for their expertise in areas for which I have limited knowledge

Finally I would like to thank my CSIRO colleagues and others, too numerous to mention, who have helped me along the way with their support and expertise.

I would like to dedicate this thesis to my children in the hope that they pursue their thirst for knowledge to make the world a better place - no matter how old they may be.

ABSTRACT

Calcrete has been shown to contain significant Au, derived from nearby mineralisation, and this has led to its current use as an exploration sampling medium. Calcretes are secondary carbonates, principally consisting of calcite and dolomite that may precipitate in regolith in a semi-arid climate. They overprint existing regolith material and commonly contain this material (e.g. soil, colluvium, laterite, saprolite and rock) on, and within, which they form. Vadose (pedogenic) and phreatic (formed by groundwater) are the two principal types of calcrete. Pedogenic calcretes are those that form in unsaturated soil horizons and are widely distributed in southern Australia and in all continents of the world. They may dissolve and re-precipitate within soil horizons, giving rise to several generations of carbonates, depending on changing climate conditions. This thesis will be restricted to pedogenic calcretes.

Despite the common use of pedogenic calcrete as a sampling medium there is a paucity of published research on how Au anomalies actually form in this material. Thus fundamental research is required to be undertaken on the nature of the Au-in-calcrete association, which will promote better understanding of how, when and where calcrete sampling is applicable for the exploration industry. In order to investigate the relationship between Au and calcrete several Au prospects from South Australia were selected for field and laboratory study, including Challenger, Barns, and Edoldeh Tank. Additional samples were investigated from the Bounty Deposit (Western Australia) where Au concentrations in calcrete are an order of magnitude higher than the South Australian counterparts.

At Challenger, the origin of the calcrete that is associated with Au was investigated. Pre-Cambrian rocks, such as the Archaean at Challenger, have very high $^{87}\text{Sr}/^{86}\text{Sr}$ ratios compared to rocks that have only recently been formed such as those derived from marine sources. Strontium isotopes were determined at Challenger and demonstrated to be derived from a marine source rather than the bedrock, despite the association of Sr (and, by inference, Ca) with Au. It was concluded that pedogenic processes (e.g. capillarity, bioturbation, evapotranspiration) caused exotic alkaline earth metals to become associated with residual Au. At Challenger, more than 95% of the Sr (Ca) was derived from marine sources (dust, rainfall and/or aerosol). The relationship between marine-

derived calcrete and Au has never been established for auriferous calcrete before and is significant since it suggests that the origin of the alkaline earth metals (Sr and Ca) is not important for its association with Au; Au is clearly originally derived from the regolith underlying the calcrete. Furthermore, a strong correlation with distance from the coast showed that the nearer to the ocean then the greater the contribution of marine Sr (low $^{87}\text{Sr}/^{86}\text{Sr}$ ratio) occurs. In a companion study, stable C isotopes were examined from the same auriferous calcrete samples and it was demonstrated, for the first time, that both C3 and C4 plants (trees and grasses) were equally dominant contributors to the inorganic C in the calcrete. Thus, the calcite in the soil (Ca and carbonate ion) is derived principally from marine and plant sources.

At Barns, disseminated Au mineralisation has created a contiguous Au-in-calcrete anomaly developed immediately above and within the saprolite. However, linear (seif) sand dunes have buried the anomaly in places. A biogeochemical survey was undertaken along and across the sand dunes over mineralisation at Barns to examine the role of vegetation in creating the Au-in-calcrete anomaly. A clear biogeochemical anomaly was identified in plant foliage, bark and litter demonstrating that Au was being taken up by roots that were tapping into buried calcrete, or mineralisation beneath it, in some cases, at least 8 m below the surface, and then depositing the Au at the surface on top of the dune. Having demonstrated the uptake of Au by plants, a dune was excavated and powdery calcrete developed in a rhizosphere within the dune was investigated. Significantly, the calcrete was shown to be highly anomalous in Au (five times above background) and thus, for the first time, it was shown that plants have had a key role in the development of a Au-in-calcrete anomaly, and, importantly, in transported regolith. Furthermore, thermoluminescence of quartz grains was commissioned and shown that the emplacement of sand above the calcrete was about 26 000 years old, indicating that the anomaly was younger than this. Mass balance calculations were performed and showed that the Au in the sand dune may have accumulated in less than 10 000 years; calcrete around the root has developed during the life of the tree. Clearly, Au-in-calcrete anomalies can form relatively rapidly compared with the age of sediments themselves.

As Au concentrations were generally low (<20 ppb) at Barns, the nature of the Au-in-calcrete was investigated in samples from Bounty where concentrations reach 1 ppm. Having established a causal relationship between Au-in-calcrete and vegetation at Barns, direct evidence of the role of plants at the micron scale was sought. Samples were chosen

from a soil profile that showed a strong positive correlation between Au and Ca and thus a minimal effect of detrital Au. Using LA-ICP-MS, SXRF (synchrotron X-ray fluorescence) and XANES (micro-X-ray absorption near-edge structure), it was shown for the first time that (i) the distribution of Au-in-calcrete at the micron scale was variable and (ii) Au occurs in both particulate and ionic form. Furthermore, ionic Au associated with Br was found in a root tubule. These observations are evidence of an evapotranspiration model for the formation of the strong, down-profile relationship between Au and calcrete i.e. Au has been mobilised then precipitated with the carbonate as vadose water has been removed from the soil by vegetation. Bromine is a typical element (with chloride and sulphate) found in evaporates. There is no chemical affinity between Au and Ca in the soil profile since further detailed analyses on sub-samples using wet chemical, LA-ICP-MS (laser ablation inductively coupled mass spectrometry) and SXRF techniques show that a Au-Ca relationship is not apparent at the sub-millimetre scale. Gold and Ca are behaving similarly but independently and they do not (at the μm scale) form a chemical bond with carbonate minerals.

At Edoldeh Tank (ET) Au prospect the distribution of Au on a tenement-scale was investigated. It was shown that geomorphological factors influence the shape and tenor of Au-in-calcrete anomalies. By their very nature Au-in-calcrete anomalies are dispersed over a wide area (to make them an effective sample medium) and this study served to document the factors involved. The ET prospect (Gawler Craton, South Australia) hosts one of the largest Au-in-calcrete anomalies in Australia and is typical of many such prospects identified in the region. For *in situ* regolith, Au is concentrated in surficial calcrete and above an upper saprolite zone depleted in Au. The Au anomaly extends downslope from a ridge of calcrete and silcrete covered saprolite into adjacent and transported regolith dominated by thin (~5 m) aeolian sand cover. The anomaly is locally broadened by downslope hydromorphic dispersion and cemented within laminated calcrete in the transported regolith. The laminated calcrete was examined in detail and the nature of the Au was found to be similar to that found at Bounty i.e. the Au in the calcrete occurs in nano-scale particulate and possibly ionic forms. In field experimental studies indicate that Au and also Ag are actively dispersing in the soil. The Ag content may be a means of distinguishing transported anomalies from those developed *in situ* and the calcrete is much older (120 000 years) than that found at Barns.

The combined individual site studies at Challenger, Barns, ET and samples from Bounty advance our understanding of the formation of Au-in-calcrete anomalies. The work has shown that an association between Au and calcrete can form at the sub-micron (ionic) to profile scale from both abiotic (geomorphology, climate, mineralisation style) and biotic (vegetation) factors; abiotic factors can shape the overall form, tenor and evolution of the Au-in-calcrete anomaly. No direct evidence was found of the role that micro-organisms play in the formation of Au-in-calcrete anomalies although experimental results from elsewhere suggest that this may be plausible, at least, in the laboratory. Further work on the role of bacteria on the formation of Au-in-calcrete anomalies in the natural environment should be encouraged. The size and shape of Au-in-calcrete anomalies may be influenced by mineralisation style, hydrology and topography and while these were briefly investigated further work is needed.

TABLE OF CONTENTS

CHAPTER 1	9
1 Preamble	9
2 Nomenclature of calcrete	9
3 Calcrete classification	11
4 Carbonate distribution	11
5 The origin of calcrete	12
6 Mineral exploration case histories	13
7 Objectives of research	16
8 Research Design	16
9 Organisation of thesis	19
10 Co-authorship of papers	21
CHAPTER 2	25
CHAPTER 3	27
CHAPTER 4	29
CHAPTER 5	31
CHAPTER 6	32
CHAPTER 7	34
GENERAL DISCUSSION	34
CHAPTER 8	39
CONCLUSION	39

CHAPTER 1

INTRODUCTION

1 Preamble

An association of Au with calcrete or soil carbonate has been known about for over a half of century but has only been documented in detail and exploited by industry since the late 1980's. The initial discovery and documentation of the strong Au-in-calcrete relationship was from the Bounty Gold Deposit (Western Australia) in 1987 (Lintern, 1989). Calcrete is an easily identifiable and common soil material found in arid to semi-arid parts of the world, including southern Australia (Goudie, 1972). Gold accumulates in calcrete and particularly in areas closer to mineralisation (Lintern and Butt, 1993a). Thus, the association is of great economic importance since its use as a geochemical sample medium will assist exploring for buried Au deposits. Many Au prospects have been discovered over the last two decades using calcrete (e.g. Drown, 2003) with some being developed into mines (Edgecombe, 1997) and this success continues. Despite the discovery of the Au-calcrete association, and clear financial incentive, detailed knowledge on the process and factors involved has remained limited. A better understanding of biotic and non-biotic factors that influence the way Au anomalies form in calcrete may improve our understanding of the process, improve mineral exploration techniques and reduce exploration costs. In order to study Au-in-calcrete anomalies an appreciation of calcrete itself is required; this includes its nomenclature, classification, distribution and origins.

2 Nomenclature of calcrete

There are a number of terms that describe the carbonate-rich material that occurs in the regolith and include calcrete, pedogenic carbonate, regolith carbonate accumulations, soil carbonate, soil lime, and soil inorganic carbon. Common alternative terms for calcrete include caliche (N. America), kunkar (India), croute (costra, carapace,) calcaire (France); other terms are akkyrshi, bhata, chamara gota, cornstone, calcin, canto (tosca) blanco, chebi-chebi, dhandla, deckkalk, gitti, harsua, kafkalla, kalk kruste, mbuga limestone, nari, patee, rimrock, reh, sabath, torba beda, tafezza, trab, trfkert, tepetate and vlei limestone (Blake, 1902; Goudie, 1972; Lamplugh, 1902; Roy, 2009). The multitude of local names for calcrete reflects its importance to local communities where it has found important use principally as a building and road making material.

“Calcrete” is essentially a field term that commonly occurs in the literature. The term “calcrete” has been used by mineral exploration companies and others since its brevity and historical use gives it advantages over terms such as “pedogenic carbonate”, “regolith carbonate accumulations” and “soil carbonate”; for similar reasons the term “caliche” is used in North America. One of the disadvantages of using the term “calcrete” is that it has connotations of being indurated as with the term “concrete” and this has led to some confusion as to its identification in the field.

The term calcrete was first suggested by Lamplugh, (1902) for a conglomerate (near Dublin, Ireland) consisting of surficial sand and gravel cemented into a hard mass by calcium carbonate precipitated from solution and redeposited through the agency of infiltrating waters, or deposited by the escape of carbon dioxide from vadose water. “Caliche” was introduced into the literature about the same time and refers to a calcareous pedogenic horizon(s) (Blake, 1902). A modern definition of calcrete describes “regolith carbonate accumulations, forming more or less well cemented aggregates composed largely of calcium carbonate, but not excluding dolomitic or magnesian material” (Eggleton, 2001). It includes massive, pisolitic, pebbly, laminar (slabby) or powdery forms that respond positively to the 0.1M HCl test by producing CO₂ gas. This modified definition is more appropriate for this thesis and thus will be used to describe these accumulations of calcite, dolomite or other carbonates in soil.

Groundwater (valley or phreatic) calcrete or those formed on marine limestone rocks have formed in a different way and will not be discussed in this thesis. Groundwater calcretes are precipitated from groundwater and typically form in the axis of dry river valleys. They can be many tens of kilometres in length and are important for U mineralisation (Carlisle, 1980; Deutscher et al., 1980). Gold occurrences have been reported in groundwater calcretes from Western Australia (Ypma, 1991). Calcrete is also distinct from limestone rock which is a sedimentary unit derived from marine deposition and commonly contains marine shells. However, calcrete may develop on the surface of limestone and/or be derived from it. Thus any distinction between limestone rock and calcrete which has been pedogenically derived from it at the surface is imprecise. Much of South Australia is covered in calcrete which has some constituents that were probably originally derived from aeolian products of weathering limestone rocks of the Nullarbor Plain (Crocker, 1946). Even inland calcrete may contain identifiable casts of marine organisms of apparent aeolian origin (e.g. foraminifera tests, Sheard, 2007).

3 Calcrete classification

Many attempts have been made to classify calcrete types (e.g. Goudie, 1983; Netterberg, 1980; Carlisle, 1980; Van Zuidam, 1976). The classification systems are based on colour, texture, carbonate crystallinity, gross morphology, hardness, hydraulic setting, degree of maturity, mineralogy, geochemical composition, biogenic features, genesis or other systems. The most common and practical systems are based on morphology; these facilitate field descriptions and avoids the problem of genesis (Chen, 2002). It has been proposed that different calcrete forms dominate different landscapes within Australia (Anand et al; 1997; Chen et al., 2002). In this thesis, it was convenient to lend terms from the classification system of Netterberg (1980) including powdery, nodular, pisolitic, tubular, hardpan (including laminated calcrete), and boulders or cobbles (massive calcrete).

4 Carbonate distribution

Calcretes are found throughout the world and in particular western USA, southern South America, southern and northern Africa, the Middle East , southern and eastern Europe, central Asia and Australia (Figure 1).

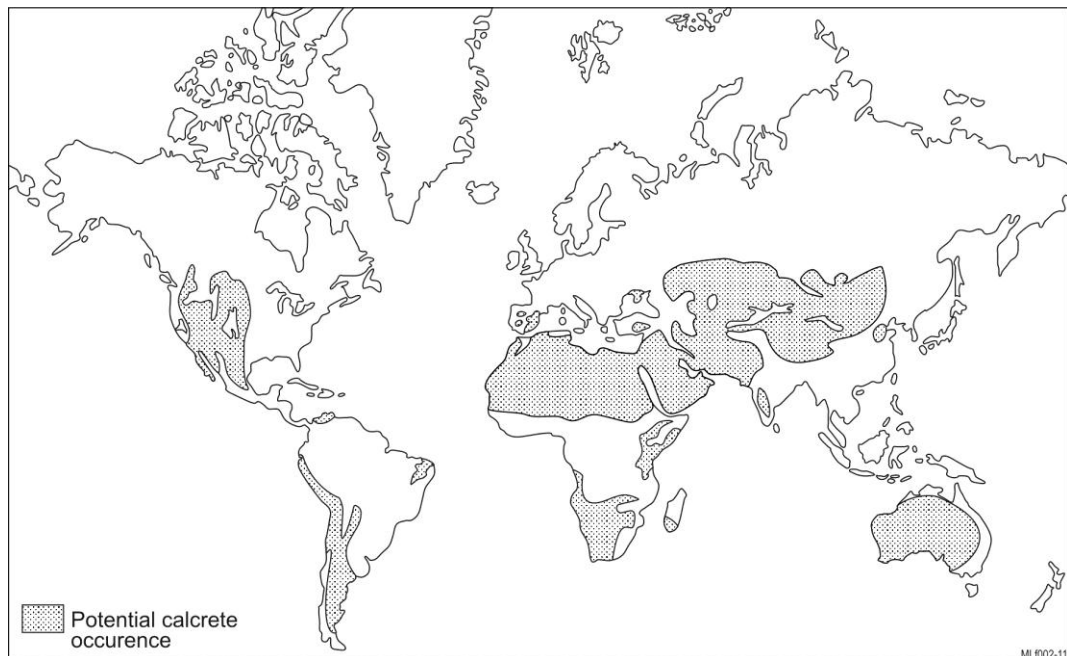


Figure 1: Distribution of probable calcrete occurrence. Derived from soil inorganic C ($>8 \text{ kg m}^{-2}$ to 1 m depth) map (modified from Lintern, 2002; data from FAO-UNESCO, 2000).

In Australia, the distribution of calcretes has been variably documented (e.g. Northcote et al., 1975; reviewed by Chen et al., 2002). In Western Australia, the Menzies Line is an important environmental and geological diffuse boundary relating to the distribution of calcrete and hence exploration techniques in the Yilgarn Craton. It is a broad (up to 100 km wide) EW transitional zone, stretching across the southern Yilgarn Craton where there are marked changes in soil types, vegetation associations and groundwater quality (Butt et al., 1977; Mahizhnan, 2004). The changes are possibly a response to climatic factors, although the sharpness with which the changes occur is more abrupt than any climatic gradient (Butt et al., 1977; Carlisle et al., 1978). South of the Menzies Line, soils are predominantly neutral to alkaline, orange to red loams, with extensive development of pedogenic calcrete. Non-calcareous earthy sand soils occupy high landscape positions, principally over granitic rocks. Groundwaters tend to be saline, neutral to acid. Average annual rainfall generally exceeds 225 mm, mainly in winter; annual evaporation is less than 2500 mm and the annual mean temperature is less than 19°C. North of the Menzies Line, soils are predominantly neutral to acid, red, non-calcareous earths, sands and lithosols, with extensive development of red-brown siliceous hardpans. Groundwater (or valley) calcretes are common in the axes of major drainages. Groundwaters are neutral to alkaline and less saline than in the south. Annual rainfall is generally less than 225 mm, falling mainly in the summer, with annual evaporation exceeding 2500 mm and annual mean temperatures exceeding 19°C.

5 The origin of calcrete

Calcretes have been described from around the world but few specifically from near or over Au deposits. Calcrete forms, and is retained, in soils that have specific environmental conditions (e.g., Lintern, 1997; Anand and Paine, 2002; Figure 2). The formation process is highly complex and involves interaction of a number of site-specific factors at a variety of scales. For example, calcrete type in a particular soil profile may be dependant on substrate (soil type; Northcote et al., 1975), rainfall (frequency, timing and volume; Jenny, 1941), evapotranspiration rates, proximity to Ca source (marine or local bedrock; Anand et al., 1997), geomorphology (Semmel, 1982) and aspect. Progressively, calcrete will replace (Nahon et al., 1977, Wang et al., 1994) and displace host materials (Watts, 1978) until the original regolith host or even bedrock becomes barely unrecognisable. Calcretes may form as a result of biological activity (e.g. Semeniuk and Searle, 1985; Philips et al., 1987; Wright et al., 1988; Alonso-Zarza, 1999; Goudie, 1996; Anand et al., 1997; Loisy et al., 1999; Zhou and Chafetz, 2009). Calcretes may be dismantled by erosion, bioturbation and local hydrological processes (Arakel, 1991).

Gold bearing calcrete needs to be characterised with the same rigour as that applied to calcrete that is not associated with Au. In addition, other factors may need to be investigated including mineralisation-style (Au particle size, Au chemistry, lithology), regolith (degree of weathering, presence of secondary cementation, landforms) and biota (vegetation, micro-organisms, bioturbation).

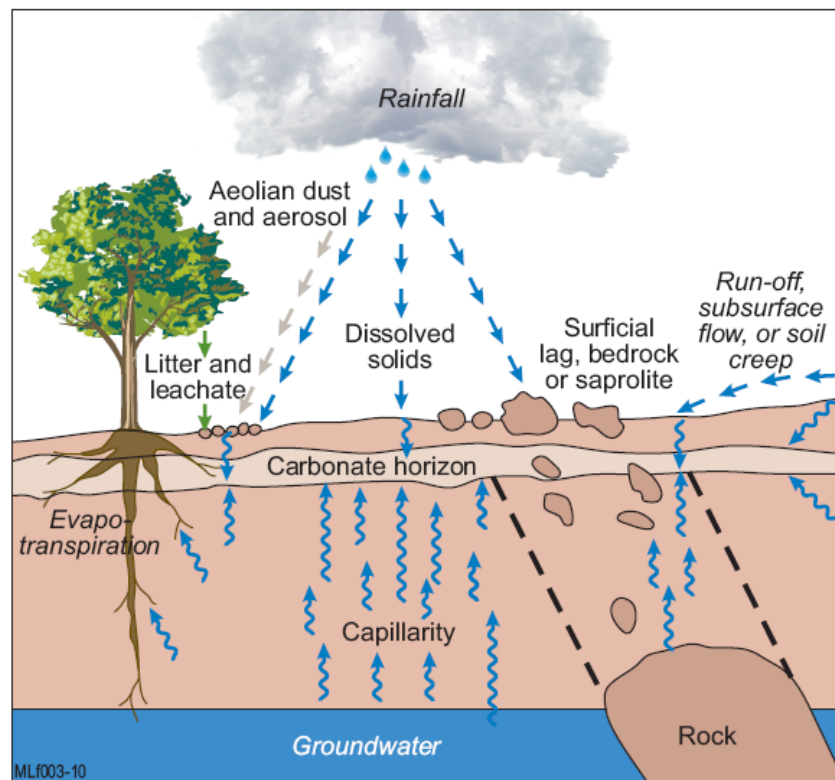


Figure 2: Schematic diagram showing possible sources of calcium in pedogenic carbonate (calcrete) horizon (Lintern, 1997).

6 Mineral exploration case histories

A historically-based literature review of the use of calcrete in mineral exploration (with an emphasis on Au) was undertaken; this comprised a chapter in a book (Lintern, 2002). In addition, literature reviews were conducted for each of the five published papers for this thesis. A brief summary of the book review follows. Calcrete sampling has had a relatively recent history as far as mineral exploration is concerned. According to McGillis (1967), the use of calcrete as a geochemical guide to metal deposits began in Russia during the late 1950s, although this did not include Au deposits. Its potential may have been recognised earlier by Cuyler (1930) who suggested Ca-rich waters, rising

under hydrostatic pressure, deposit calcrete in faulted areas, which are commonly associated with mineral deposits. Outside of Russia, the value of calcrete as a specific geochemical sampling medium was not fully recognised until the early 1970s, although early reference to a possible HCl soluble Au component in calcrete was made by Sokoloff (1949). One of the first records of calcrete as a geochemical sampling medium appears to have been in the Yilgarn Craton, Western Australia when it was investigated as a means to explore for Ni deposits. Initially, it was considered as a geochemical diluent (Mazzuchelli, 1972) and other media such as the residual soils themselves were sampled instead or efforts were made to upgrade the metal content of samples by dissolving the calcrete away and analysing the residue, (e.g. Garnett et al., 1982). However, Cox (1975) gave further consideration to its specific use when looking for Ni in the Kambalda area (Western Australia). Other mainly base metal studies in South Africa and Australia followed during the next decade (e.g. Mazzucchelli et al., 1980; Frick, 1985; Leduc, 1986; Guedria et al., 1989; Tordiffe et al., 1989; Harrison, 1990). The general Au-calcrete association has been recorded in several anecdotal accounts (e.g., Kriewaldt, 1969; K. Schulz, pers. comm. to C.R.M. Butt, 1985) and in the scientific literature (e.g., Smith and Keele, 1984; Mann, 1984a, b; Smith, 1987; Lawrance, 1988a; Glasson et al., 1988); however, it appears that some of these accounts may be referring to fortuitous detrital Au particles encapsulation in calcrete directly, or associated, for example, with ferruginous pisoliths (laterite), rather than a true correlation. Despite these studies, exploration companies have never systematically used calcrete as the sample medium of choice during these early years and it was never considered for Au exploration.

Calcrete became a specific sample medium during Au exploration in Australia from the late 1980s until the present as a result of CSIRO (Commonwealth Scientific and Industrial Research Organisation) and later CRC LEME research (Cooperative Research Centre for Landscape Evolution and Mineral Exploration). In 1987, the CSIRO commenced a research project with a consortium of exploration companies through AMIRA (Australian Mineral Industries Research Association Ltd) to improve geological and geochemical methods for mineral exploration that would facilitate the location of blind, concealed or deeply weathered Au deposits. In late 1987, the first detailed research on calcrete above a Au deposit commenced at Bounty (120 km south of Southern Cross, Western Australia) with spectacular results (Lintern, 1989; Lintern et al., 1992; Lintern and Butt, 1993a). For the first time a precious (apparently inert) metal was demonstrated to be highly correlated (not merely associated) with Ca (calcrete), a mobile element in the

soil; this was strong evidence for the soluble and mobile nature of Au in the soil. The results also showed not unexpectedly that Fe and many other elements had been diluted by the calcrete, consistent with the earlier base metal studies, and so were not correlated with Ca (Lintern and Butt, 1993a). The correlation was widespread at the Bounty Deposit in soils typical of those found throughout the auriferous greenstones of the Eastern Goldfields of Western Australia. Thus, there was a great potential for the widespread use of calcareous soil (calcrete) for exploration purposes. Further research confirmed the relationship to be robust throughout the southern Yilgarn Craton and indicated that calcrete was a geochemical sample medium that could be used with confidence, pending certain important provisos (Lintern et al., 1996, 1997).

Calcrete sampling reached a significant milestone as an exploration technique when its use was publicly acknowledged to have located the Challenger Gold Deposit (Gawler Craton, South Australia) in 1995. Subsequently, in a series of conference proceedings, company reports, magazine reviews and newspaper articles describing Au exploration in the Gawler Craton, calcrete was highlighted and recommended by many as the principal sampling technique to be used (e.g. Bonwick, 1997; Edgecombe, 1997). The success of calcrete sampling in SA subsequently spread into NSW (Smith et al., 1996; Hill et al., 1998; McQueen, 2006) and Victoria (Anon, 1998).

By the end of the 1990s, calcrete was being acclaimed as the sampling medium of choice back in the Yilgarn Craton by a much wider audience of exploration companies than the early 1990s, many of whom were not familiar with the earlier Western Australian research, but had been swayed by the successes in SA (e.g. Rubicon, Hornet and Pegasus discoveries, Boyer and Grivas, 1999; Golden Cities discovery, Kehal et al., 1999; Ghost Crab discovery, Miller et al., 1999). Clearly the technique has taken several years to filter through to the junior explorers, despite the new discoveries being located in the Yilgarn where it was first recognised. As with many other geochemical sampling media, such as soil, lateritic residuum, rock chip or stream sediments, the popularity of calcrete has benefited from analytical laboratories providing low-cost, rapid chemical analyses with low detection limits. Calcrete is used in many other parts of the world for mineral exploration including the Americas (S. Gatehouse, pers. comm; L. Bettenay, pers. comm.; Anon, 1998), southern and northern Africa, (Frick, 1985; Leduc, 1986; Guedria et al., 1989; Tordiffe et al., 1989), and parts of the former USSR. Recently, calcretes were used in the exploration for kimberlites (Roy, 2009).

7 Objectives of research

The principal research question to be answered here is *what are the factors involved in Au-in-calcrete anomaly formation?* This may involve a number of subsidiary questions including some that may be beyond the scope of this thesis:

- Where is the Au located in the calcrete?
- Are there any mineralogical associations of Au-in-calcrete?
- Under what conditions will Au be precipitated in/on and mobilised from calcrete?
- Do biologically-generated ligands mobilise Au in calcareous soils or is it a purely inorganic process?
- How long does it take to form Au-in-calcrete soil anomalies?
- What is the principal mechanism by which Au-in-calcrete anomalies form?
- Do the mechanisms of Au-in-calcrete formation differ depending on site conditions?

Thus, the principal objective of this research is to better understand the formation of Au-in-calcrete anomalies in the regolith and, based on this, develop more effective procedures for their use in mineral deposit detection. Specific objectives to answer some of the research questions include:

- Determining the geochronology of selected Au-in-calcrete anomalies.
- Determining the origin of Au-bearing calcrete.
- Establishing the nature of Au contained within the calcrete.
- Characterising calcareous soil profiles that contain Au.
- Determining controls on the formation and evolution of Au-in-calcrete anomalies.

Research on Au-in-calcrete to date has largely been involved with documentation of the phenomenon and determining its spatial extent for exploration purposes. The main problem is that few process studies have been undertaken and so little is known about the detailed nature of Au-in-calcrete and how the Au actually resides in the calcrete.

8 Research Design

The research design for this thesis followed the plan as set out in the candidacy. The plan was to review gold in calcrete occurrences, then investigate the fundamental question of whether calcretes associated with gold deposits e.g. at Challenger were essentially any

different from other calcretes. This completed the plan was then to examine broad scale influences on the distribution of gold in calcrete e.g. at ET before investigating specifically the compelling control that vegetation has on gold in calcrete at the soil profile level. Following this, detailed examination of samples to look at possible biological influences was undertaken. The research flow did not necessarily occur in that precise order as avenues of research opened at specific sites as the study evolved.

The research was conducted at four principal sites in Australia (Figure 3), three in South Australia and one in Western Australia. All sites contained calcrete but varied in other site characteristics such as style and extent of mineralisation, regolith thickness and type, vegetation, climate and groundwater (Table 1).

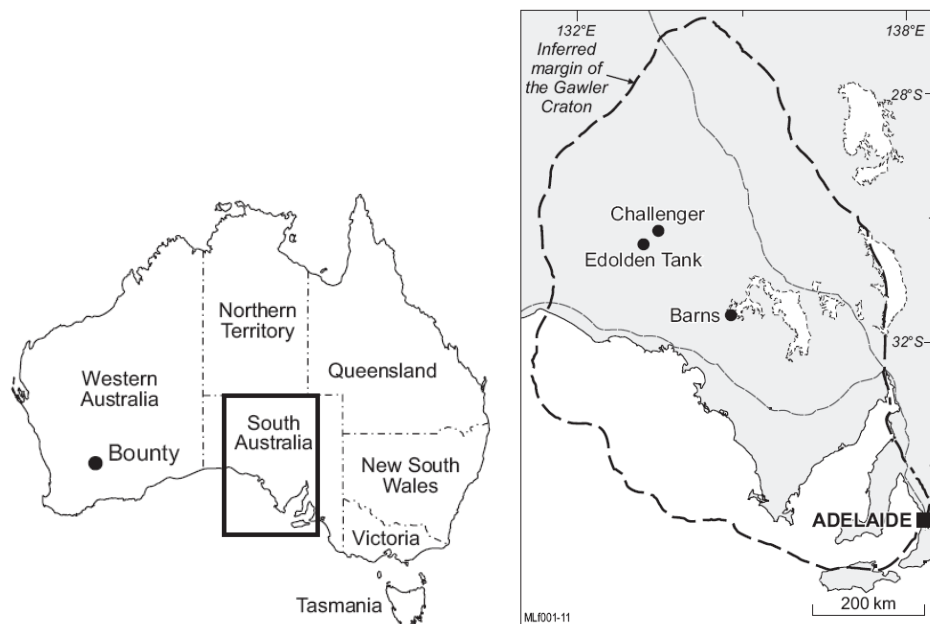


Figure 3: Diagram showing locations of main study sites where research was conducted for this thesis.

Table 1: Characteristics of the principal sites accessed in this study.

Characteristics	Challenger/ Gawler Craton	Edoldeh Tank	Barns	Bounty
Thickness of regolith	> 50 m	> 50 m	> 50 m	> 50 m
Thickness of transported cover	1 m	5 m	8 m	2 m
Depth to mineralisation	3 m	30 m	30 m	5 m
Vegetation	Bluebush Senna	Acacia Eucalyptus Casuarina	Eucalyptus	Eucalyptus

Soil type	Red-brown hardpan	Sandy clay	Sand	Heavy clay
Rainfall				
Groundwater depth	Not found	50 m	35 m	Not found

The investigations were multi-disciplinary and required specialist assistance from some other researchers e.g. synchrotron analyses. Gold anomalies were selected and mapped according to site characteristics. Calcrete anomalies were studied with possible mitigating factors including calcrete type, environmental factors and the mineralisation-style they are derived from. Soil profiles and soil samples were studied in detail to identify their chemical, mineralogical and biological characteristics using the following techniques: laser ablation inductively coupled mass spectroscopy (LA-ICPMS), ICPMS, ICP optical emission spectroscopy (ICPOES), X-ray diffraction (XRD), synchrotron X-ray fluorescence (SXRF) and ion microprobe. Samples were collected to conduct laboratory tests on solubility and nature of Au complexes in soil. The role of biota in Au anomaly formation was critically examined. The age of Au-in-calcrete anomalies was also determined to get a better understanding on how quickly they may form. The work conducted at each principal site is summarised in Table 2.

Table 2: Studies conducted at the four principal research sites.

Study conducted	Challenger/ Gawler Craton	Edoldeh Tank	Barns	Bounty
Regolith characterisation	yes	yes	yes	yes
SEM	yes	yes	no	yes
Synchrotron	no	yes	no	yes
Laser ablation ICPMS	no	yes	yes	yes
Isotopes (Sr and C)	yes	yes	yes	no
Thermoluminescence dating	no	yes	yes	no
Biogeochemistry	yes	yes	yes	yes
Microbiology	no	yes	no	yes

This research contributes towards answering some of the many questions that remain for understanding this important phenomenon and will ultimately assist Au exploration companies in their quest for the discovery of new ore deposits.

9 Organisation of thesis

This thesis is comprised of eight chapters. Chapter 1 (this chapter) is an introduction and includes an explanatory overview of the work, significant findings of the thesis, describes how the research papers are linked, identifies the limitations of the research and highlights future directions. A brief literature survey is described which is an extract from the full literature survey which was published as a book chapter and is reproduced in Appendix 1 (Lintern 2002). Chapter 2 to 6 are Microsoft Word™ versions of papers that have been published as pdf documents in international journals (Table 3).

Chapter 2 describes the source of the carbonate and alkaline earth metals contained within pedogenic carbonate containing Au at the Challenger Gold Deposit. It also demonstrates the strong marine influence on pedogenic carbonates in an east-west transect to the east of the Nullarbor Plain.

Chapter 3 details the preliminary study at Edoldeh Tank gold anomaly in the Western Gawler Craton. The influence of geomorphology in shaping geochemical anomalies is shown.

Chapter 4 is concerned with experimental work at part of the Edoldeh Tank gold anomaly and shows that Au is being mobilised as an ionic species in the soil.

Chapter 5 clearly shows that vegetation is playing a strong role in the formation of gold in calcrete anomalies at Barns gold prospect.

Chapter 6 details laboratory analyses of calcrete and shows at the micron scale how biota are playing a role in anomaly formation in the rhizosphere and that Au occurs partly in an ionic form and related to evaporation.

Chapter 7 discusses the findings from the thesis and shows how the studies from the different sites are linked and show that both biotic and abiotic factors are involved in Au-in-calcrete formation.

Chapter 8 concludes the thesis.

Table 3: The published and accepted papers written for this thesis.

Title of paper or book chapter	Authorship	Publication details	Journal reviewers	Chapter in thesis
Calcrete sampling for mineral exploration	M.J. Lintern	In Calcrete: characteristics, distribution and use in mineral exploration (eds. X.Y. Chen, M.J. Lintern and I.C. Roach). CRC LEME, Perth, Australia, 31-109. (2002)	M. Skwarnecki. M. Cornelius C. Butt R. Anand	Summary in 1
The source of pedogenic carbonate associated with Au-calcrete anomalies in the western Gawler Craton, South Australia	M.J. Lintern M.J. Sheard A.R. Chivas	Chemical Geology 235 (2006): 299–324	D. Rickard Anonymous reviewers	2
The gold-in-calcrete anomaly at the ET gold prospect, Gawler Craton, South Australia	M.J. Lintern M.J. Sheard N.B. Buller	Applied Geochemistry 26 (2011): 2027-2043	D. Arne M. Arundell Anonymous reviewers	3
Experimental studies on the gold-in-calcrete anomaly at Edoldeh Tank gold prospect, Gawler Craton, South Australia	M.J. Lintern R.M. Hough C.G. Ryan	Journal of Geochemical Exploration 112 (2012) 189–205	R. Koole A. Schmidt-Mumm B. Smee Anonymous reviewers	4
Vegetation controls on the formation of Au anomalies in calcrete and other materials at the Barns Gold Prospect, Eyre Peninsula, South Australia	M.J. Lintern	Geochemistry: Exploration, Environment, Analysis, 7 (2007): 249–266	G.E.M. Hall C.E. Dunn Cliff Stanley	5
Ionic Au-in-calcrete revealed by LA-ICP-MS, SXRF and XANES	M.J. Lintern R.M. Hough C.G. Ryan J. Watling M. Verrall	Geochimica et Cosmochimica Acta 73 (2009): 1666-1683	J. Chorover Anonymous reviewers	6

10 Co-authorship of papers

The paper entitled “The source of pedogenic carbonate associated with Au-calcrete anomalies in the western Gawler Craton, South Australia” included the work of two co-authors whose contributions are outlined below.

Melvyn Lintern

Responsible for the collection of samples. Overall writing and compilation of the manuscript, including both scientific and grammatical editing. Responsible for the overall generation and interpretation of scientific data including that from Challenger, the east west transect and other areas from the western Gawler Craton. Responsible for the generation of Figures 1-3, 5-15.

Malcolm Sheard

Assistance with choice of sample collection sites. Largely responsible for descriptions under “Calcrete” in “Study locations” and for descriptions in Table 1. Scientific and grammatical editing. Responsible for the generation of Figure 4.

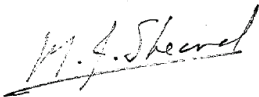
Allan Chivas

Scientific and grammatical editing. Responsible for most of the isotopic analyses.

Signed:



Melvyn Lintern



Malcolm Sheard



Allan Chivas

The paper entitled “Ionic Au-in-calcrete revealed by LA-ICP-MS and synchrotron radiation” included the work of four co-authors whose contributions are outlined below.

Melvyn Lintern

Responsible for the initiation of ideas for experiment and collection of samples. Responsible for the overall conducting of scientific experiments. Overall writing and compilation of the manuscript, interpretation of processed data, and scientific and grammatical editing. Responsible for the overall generation of scientific data.

Rob Hough

Assisted with operation of synchrotron experiments and production of XANES spectra from the synchrotron. Writing of some of the synchrotron methods section Some grammatical editing.

Chris Ryan

Responsible for the operation of the synchrotron, processing of raw data and generation of GeoPIXE geochemical maps. Writing of some of the synchrotron methods section. Some scientific editing.

John Watling

Assisted with initial discussions of LA-ICP-MS experimental design. Some grammatical editing.

Michael Verral

Assisted with operation of SEM, elemental analyses using EDAX and production of SEM images. Some grammatical editing.

Signed:



Melvyn Lintern



Rob Hough



Chris Ryan



John Watling



Michael Verral

The paper entitled “The gold-in-calcrete anomaly at the ET gold prospect, Gawler Craton, South Australia.” included the work of two co-authors whose contributions are outlined below.

Melvyn Lintern

Responsible for the initiation of the work. Responsible for the collection of samples. Overall writing and compilation of the manuscript, including both scientific and grammatical editing. Responsible for the overall generation and interpretation of scientific data. Responsible for the generation of all figures.

Malcolm Sheard

Largely responsible for descriptions under “Calcrete” in “Study locations” and for descriptions in Table 1. Minor scientific and grammatical editing.

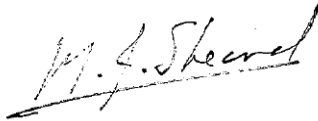
Nicola Buller

Assisted with microbiological methodology. Minor scientific editing.

Signed:

A handwritten signature in black ink, appearing to read 'M. Lintern', with a large loop at the bottom.

Melvyn Lintern

A handwritten signature in black ink, appearing to read 'M. Sheard', with a horizontal line underneath.

Malcolm Sheard

A handwritten signature in black ink, appearing to read 'N. Buller', with a horizontal line underneath.

Nicola Buller

The paper entitled “Experimental studies on the gold-in-calcrete anomaly at Edoldeh Tank gold prospect, Gawler Craton, South Australia” included the work of two co-authors whose contributions are outlined below.

Melvyn Lintern

Responsible for the initiation of the work. Responsible for the collection of samples. Overall writing and compilation of the manuscript, including both scientific and grammatical editing. Responsible for the overall generation and interpretation of scientific data. Responsible for the generation of all figures.

Rob Hough

Assisted with operation of synchrotron experiments. Generation of distribution images of using GeoPixe for Figure 16 (Lintern et al., in press).

Chris Ryan

Responsible for the operation of the synchrotron and collection of raw data. Minor editing in Synchrotron methods section.

Signed:

A handwritten signature in black ink, consisting of a series of vertical, slightly curved strokes that form a dense, somewhat rectangular shape, with a small horizontal stroke at the bottom.

Melvyn Lintern

A handwritten signature in black ink, featuring a series of connected, wavy, horizontal strokes that form a continuous, somewhat elongated shape.

Rob Hough

A handwritten signature in blue ink, consisting of a few simple, connected strokes that form a cursive, somewhat elongated shape.

Chris Ryan

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CHAPTER 2

THE SOURCE OF PEDOGENIC CARBONATE ASSOCIATED WITH GOLD-CALCRETE ANOMALIES IN THE WESTERN GAWLER CRATON, SOUTH AUSTRALIA

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ABSTRACT

Elevated Au concentrations above bedrock mineralisation are commonly closely associated with calcrete in arid residual and semi-residual soils in Australia. The origin of Australian calcrete has been argued for many years but there have been very few published studies. Calcrete from Au deposits and prospects and elsewhere in the Gawler Craton (South Australia) were studied for their geochemical composition, and Sr and C isotope ratios. By comparing carbonate $^{87}\text{Sr}/^{86}\text{Sr}$ ratios with underlying weathered rock and bedrock, it is demonstrated that many samples have overwhelmingly atmospheric Sr (>94%) and, by inference, Ca (>98%) rather than a local bedrock or soil mineral origin. The isotopic composition of calcretes at the Challenger Gold Deposit lie on a mixing trend of decreasingly marine Sr from the Nullarbor Plain to Tarcoola, 300 km to the east. The $\delta^{13}\text{C}_{\text{PDB}}$ values (-7.5‰ to +0.1‰) suggest that the carbonate C in calcrete has a mixed origin derived from C3 and C4 plants. A model is presented to explain how the Au–Ca association may form in calcrete at Challenger based on these new Sr and C isotope data and in which the role of plants and atmospherically deposited Sr and Ca combine with pre-existing Au in the soil. The implication for mineral exploration is that, since most of the Ca is derived from marine sources, it is not a pre-requisite that autochthonous soil minerals (derived from underlying weathered bedrock) supply Ca for calcrete to form and for the Au–Ca association to occur. Thus, the independence of the Ca

with respect to underlying lithology reaffirms the usefulness of calcrete as a sampling medium for Au exploration

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CHAPTER 3

THE GOLD-IN-CALCRETE ANOMALY AT THE ET GOLD PROSPECT, GAWLER CRATON, SOUTH AUSTRALIA

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ABSTRACT

Much of Australia has an extensive regolith cover that conceals basement rocks and hinders mineral exploration and this situation is particularly acute in the western Gawler Craton (South Australia), where, in addition to fluvial, marine and colluvial sediments, the land surface is extensively cloaked by sand dunes. This study documents the Au distribution at the ET Au prospect (Great Victoria Desert) in the western Gawler Craton (South Australia). Although no economic Au mineralisation has yet been found at ET, the prospect hosts one of the larger Au-in-calcrete anomalies in Australia and is typical of many such prospects identified in the region. In addition to calcrete, the distribution of Au in regolith and biotic sample media was also examined.

The study at ET shows that:

- (i) For *in situ* regolith, Au is concentrated in surficial calcrete and above an upper saprolite zone depleted in Au.
- (ii) The Au anomaly extends from a ridge of weathered Archaean basement into adjacent and transported regolith dominated by thin (~5 m) aeolian sand cover. The anomaly appears to be locally broadened by lateral dispersion in the transported

overburden, or, possibly, Au additions to the surface from underlying buried mineralisation.

- (iii) Calcrete appears to be the most consistent sample medium for Au providing coherent anomalies compared with soil, vegetation, near surface drill cuttings and *Bacillus cereus*.
- (iv) Gold appears to be the best target element in upper regolith or calcrete, although As may provide supplementary information on the location of prospective mineralisation.

Calcrete sampling has been a successful exploration technique to reveal cohesive Au anomalies within *in situ* regolith. Where transported regolith dominates and landforms are favourable e.g. sloping, calcrete (containing Au) will disperse (in solution and/or mechanically eroding) and thus provide a spatially larger target area for mineral exploration purposes. However, in these settings, the actual source of mineralisation may be difficult to locate due to various factors related to the dispersion processes and weathering history. At ET, other sampling media such as bacteria (*Bacillus cereus*) and vegetation have a greater uncertainty associated with them and their anomalies are not as cohesive.

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CHAPTER 4

EXPERIMENTAL STUDIES ON THE GOLD-IN-CALCRETE ANOMALY AT EDOLDEH TANK GOLD PROSPECT, GAWLER CRATON, SOUTH AUSTRALIA.

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ABSTRACT

Calcrete sampling is the near-surface exploration method of choice for Au in many drier parts of the world, particularly southern Australia. Edoldeh Tank is a weakly mineralised Au prospect in calcrete terrain that lies at the eastern edge of the Great Victoria Desert dunefield (South Australia). At Edoldeh Tank a variety of calcretes occur but the dominant form is a laminated calcrete horizon (LCH). The mineralogy of the near-surface soil is relatively simple and consists of calcite, dolomite, quartz, kaolinite and minor smectite; quartz dominates the unconsolidated overlying sandy soil, and carbonate minerals dominate the LCH. We determine the distribution and nature of the Au at a small scale using a variety of techniques, including SEM, LAICPMS and SXRF and dated sediments to understand calcrete genesis. In a series of thirty excavated soil pits, Ca and Au concentrations increased with depth, markedly so at the LCH. We provide multiple lines of evidence to show there is a general association of Au with calcrete but not a strong correlation as seen with soil profiles elsewhere that have younger, recently formed powdery calcrete. Experiments suggest Au and Ag are currently mobile in this environment despite the low rainfall and that Au occurs in two forms: Au (possibly ionic) occurs throughout the sample with some regions having higher concentrations than others; particulate Au occurs randomly but is more common where the general level of Au is higher. The laminated nature of the calcrete suggests it has formed episodically.

An association of Ag with Au in calcrete suggests a means to distinguish anomalies that have developed in residual regolith from those that have dispersed into adjacent sediments. Laminated calcrete is just as effective an exploration sample medium as powdery calcrete. Mobilised Ca, Au and Ag in calcrete can extend the lateral extent and distance from the source of the geochemical anomaly thus providing an effective vector to target for sampling. A landscape dispersion model of Au in calcrete is presented, which requires further testing, to assist the mineral explorer in covered terrains.

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CHAPTER 5

VEGETATION CONTROLS ON THE FORMATION OF GOLD ANOMALIES IN CALCRETE AND OTHER MATERIALS AT THE BARNES GOLD PROSPECT, EYRE PENINSULA, SOUTH AUSTRALIA

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ABSTRACT

A biogeochemical study was undertaken at Barnes Gold Prospect, a Au-in-calcrete discovery in the northern Eyre Peninsula (South Australia). The prospect is located in highly weathered Proterozoic rocks and is overlain by more than a 1 m of aeolian quartz sand but, significantly, the cover thickens to 8 m over part of the mineralization due to a longitudinal sand dune. The dune is well-vegetated, with *Melaleuca* shrubs and *Eucalyptus* trees up to 5 m high.

Results indicate that anomalous Au occurred over mineralization in plant organs, litter, soils and sand. The highest Au concentrations (9 ppb) occurred in calcareous rhizomorphs high up in the dune. Luminescence dating determined that the dune took no longer than 27000 years to form and mass balance calculations indicate that the Au anomaly in the dune could have taken less than 10000 a to form. Mechanisms for the Au accumulation in the sand are postulated but it appears that a biological process, principally involving vegetation, is the most viable.

A 200 m sample spacing of vegetation appears to be adequate for exploration of this type of deposit. Below the sand, calcrete provides a robust sampling medium. At present, due to limited knowledge of exploration methods in this type of environment, the mineral explorer must either expend significant financial resources augering through areas of sand cover to collect the buried calcrete samples, or have lower confidence that vegetation and surficial soil samples will detect mineralization.

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CHAPTER 6

IONIC GOLD IN CALCRETE REVEALED BY LA-ICP-MS, SXRF AND XANES

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ABSTRACT

Highly anomalous Au concentrations in calcrete were discovered in 1987 at the Bounty Gold Deposit, Western Australia. A strong correlation was noted between the Ca, Mg, Sr and Au in soil profiles which has not only attracted the interest of mineral explorers but also chemists, soil scientists, metallurgists and climatologists. Gold has been considered an inert element and so its strong association with the alkaline earth group of relatively mobile elements is both remarkable and intriguing. Despite widespread interest, there have been few published papers on the Au-calcrete phenomenon. Here, we present work conducted on calcareous soil samples from above the Bounty mineralisation in Western Australia, prior to mining.

Using SXRF (synchrotron X-ray fluorescence) and XANES (micro-X-ray absorption near-edge structure), we have shown for the first time the distribution of Au-in-calcrete and that it occurs in both particulate and ionic form. Much of the ionic Au associated with Br is found in a root tubule. The observations are consistent with an evapotranspiration model for the formation of Au in the calcrete; Au has been mobilised then precipitated as vadose water has been removed from the soil by trees and shrubs. While the association between Au and Ca is very strong in bulk sample analyses down the soil profile, other detailed analyses on sub-samples using wet chemical, LA-ICP-MS

(laser ablation inductively coupled mass spectrometry) and SXRF techniques show that it is not apparent at the sub-millimetre scale. This suggests that the Au and Ca are behaving similarly but independently and they do not (at the μm scale) co-precipitate with carbonate minerals.

These results corroborate other studies that suggest biotic influences can affect the mobilisation and distribution of Au in surficial materials. Water-extractable Au-in-calcrete has been reported previously and the ionic Au described in this study likely represents that soluble component. The presence of easily solubilised Au in soils has been widely discussed and exploited for mineral exploration.

CHAPTER 7

GENERAL DISCUSSION

The origin of Australian calcrete has been argued for many years but there have been very few published studies. Since Au is associated with calcrete in arid soils in Australia, it is important to determine whether the Ca, Sr and inorganic C, contained within the carbonate, is derived locally from the surrounding rocks and whether this is an important factor in the formation of Au-in-calcrete anomalies. For example, do anomalies only occur if Ca and Sr are derived from local rocks? Calcrete from Au deposits and prospects and elsewhere in the Gawler Craton (South Australia) were studied for their geochemical composition, and Sr and C isotope ratios (Chapter 2). By comparing carbonate $^{87}\text{Sr}/^{86}\text{Sr}$ ratios with underlying weathered rock and bedrock, it was demonstrated that most samples are dominated by overwhelmingly atmospheric Sr and, by inference, Ca rather than a local bedrock or soil mineral origin. Furthermore, it was shown that the isotopic composition of calcretes at the Challenger Gold Deposit lie on a mixing trend of decreasingly marine Sr from the Nullarbor Plain to Tarcoola, 300 km to the east. This implies that all calcrete in the Gawler Craton has a substantial input from marine sources. The $\delta^{13}\text{C}_{\text{PDB}}$ values (-7.5‰ to $+0.1\text{‰}$) suggest that the soil inorganic carbon in calcrete has a mixed origin derived from C3 and C4 plants. A model is presented to explain how the Au–Ca anomaly may form in calcrete at Challenger based on these new Sr and C isotope data and in which the role of plants (biotic factor) and atmospherically deposited Sr and Ca (abiotic factor) combine with pre-existing Au in the soil (Chapter 2); similar origins for the carbonate were found at Barns (Chapter 5). The implication for mineral exploration is that, since most of the Ca is derived from marine sources, it is not a prerequisite that autochthonous soil minerals (derived from underlying weathered bedrock) supply Ca for calcrete to form and for the Au–Ca association to occur. Thus, the independence of the Ca with respect to underlying lithology reaffirms the efficacy of calcrete as a sampling medium for Au exploration

A typical Au-in-calcrete anomaly and the Au distribution in other regolith materials and plants at the Edoldeh Tank (ET) prospect in the western Gawler Craton (South Australia) are described in Chapter 3. The ET prospect hosts one of the largest Au-in-calcrete anomalies in Australia and is typical of many such prospects identified in the region, most of which have not been described in the literature. To date, economic mineralisation at

ET has not been found. The distribution of Au in regolith and biotic sample media was also examined to examine their relationship with calcrete.

For the geochemical survey at ET, the principal results indicate that:

- (v) For *in situ* regolith, Au is concentrated in surficial calcrete and above an upper saprolite zone depleted in Au.
- (vi) The Au anomaly extends from a ridge of weathered Archaean basement into adjacent and transported regolith dominated by thin (~5 m) aeolian sand cover. The anomaly appears to be locally broadened by lateral dispersion in the transported regolith, or, possibly, Au additions to the surface from underlying buried mineralisation.
- (vii) Calcrete appears to be the best consistent sample medium for Au providing coherent anomalies compared with soil, vegetation, near surface drill cuttings and *Bacillus cereus*.
- (viii) Gold appears to be the best target element, although As may provide supplementary information on the location of prospective mineralisation.

In general, calcrete sampling for Au exploration is a technique that works particularly well at Edoldeh Tank and provides coherent anomalies for *in situ* regolith. Where transported regolith dominates and landforms are favourable, calcrete (containing Au) will disperse (chemically and mechanically) and thus provide a spatially larger target area for mineral exploration purposes. However, in these settings, the actual source of mineralisation may be difficult to locate due to various factors related to the dispersion processes and weathering history. At ET, other sampling media such as *Bacillus cereus* and vegetation have a greater uncertainty associated with them and their anomalies are not as coherent. There is not a clear relationship between Au-in-calcrete and biota anomalies at ET.

The detailed and experimental study at ET (Chapter 3) indicates that a variety of calcretes occur but the dominant form here is a laminated calcrete horizon (LCH). This calcrete form is common throughout the world and is in contrast to the powdery calcrete found at Bounty (Chapter 6) and in the upper part of the sand profile at Barns (Chapter 5). The mineralogy of the soil is relatively simple and consists of calcite, dolomite, quartz, kaolinite and smectite; quartz dominates the unconsolidated overlying sandy soil, and carbonate minerals dominate the LCH.

The distribution and nature of the Au was determined at a large scale at ET using a variety of techniques, including SEM, LAICPMS and SXRF (c.f. Chapter 6) and the dating of sediments, that apprised us with the antiquity of calcrete (c.f. Chapter 5) in this area, was undertaken. In a series of thirty excavated soil pits, Ca and Au concentrations increased with depth, markedly so at the LCH (Chapter 3). There appears to be a general association of Au with calcrete but not a strong correlation as seen with soil profiles elsewhere that have younger, recently formed powdery calcrete. Experiments suggest Au and Ag are currently mobile in this environment despite the low rainfall and that Au occurs in two forms: Au (possibly ionic) occurs throughout the sample with some regions have higher concentrations than others; particulate Au occurs randomly but is more common where the general level of Au is higher. The laminated nature of the calcrete (the LCH) suggests it has formed episodically. The association of Ag with Au-in-calcrete suggests a means to distinguish anomalies that have developed in residual regolith from those that have dispersed into adjacent sediments.

It was concluded that the laminated calcrete at ET is just as effective an exploration sample medium as compared with powdery calcretes found elsewhere (Chapter 6). Mobilised Ca, Au and Ag in calcrete can extend the lateral extent and distance from source of the geochemical anomaly thus providing an effective vector to target for sampling. The Au distribution at this scale is related to geomorphology rather than biological controls (Chapter 5 and 6). A dispersion model of Au-in-calcrete is presented.

Biotic factors that may influence the formation of Au-in-calcrete anomalies (Chapter 5: c.f. Chapter 6). Initially, a biogeochemical study was undertaken at the Barns Gold Prospect, a Au-in-calcrete discovery in the Northern Eyre Peninsula (South Australia). The prospect is located in highly weathered Proterozoic rocks and is overlain by at least 1 m of aeolian quartz sand that thickens to 8 m as a longitudinal sand dune over part of the mineralisation. The dune is well-vegetated, with *Melaleuca* shrubs and *Eucalyptus* trees up to 5 m high. The study showed that over mineralisation anomalous Au concentrations occur in plant organs, litter, soils and sand. The highest Au concentrations (9 ppb) in the dune occur in calcareous rhizomorphic calcrete high up within the dune. Luminescence dating shows that the dune took no longer than 27 000 years to form (compared with 120 000 years at ET; Chapter 4) and mass balance calculations indicate that the Au anomaly in the dune has formed in less than 10 000 years. Mechanisms for the Au accumulation

in the sand are postulated and it is clear that a biotic process, principally involving vegetation, is the most viable mechanism by which the Au-in-calcrete anomalies form in the dune. The carbonate has its origins in plants and marine factors as (c.f Challenger; Chapter 2). Gold concentrations were too low to investigate the distribution of Au at the micron scale as at ET (Chapter 4) and Bounty (Chapter 6).

Below the sand, more indurated calcrete provides a robust sampling medium. At present, due to limited knowledge of exploration methods in this type of environment, the mineral explorer must either expend additional financial resources augering through areas of sand cover to collect buried calcrete samples, or have lower confidence that vegetation and surface soil samples will detect mineralisation. This study is comparable to that undertaken at ET (Chapters 3 and 4) and provides the link between Au-bearing calcrete created by vegetation and the larger scale Au-in-calcrete anomalies created by geomorphological influences.

The nature of the Au found within calcrete was studied from samples from Bounty Gold Deposit (Chapter 6). Highly anomalous Au concentrations in calcrete were first discovered in 1987 at the Bounty Gold Deposit, Western Australia. A strong correlation was noted between the Ca, Mg, Sr and Au (in soil profiles) which has not only attracted the interest of mineral explorers but also chemists, soil scientists, metallurgists and climatologists. Gold has been considered an inert element and so its strong association with the alkaline earth group of relatively mobile elements is both remarkable and intriguing. Using LA-ICP-MS, SXRF (synchrotron X-ray fluorescence) and XANES (micro-X-ray absorption near-edge structure), the distribution of Au-in-calcrete was shown for the first time and that Au occurs in both particulate and ionic form (c.f. Chapter 4). Much of the ionic Au associated with Br was found in a root tubule clearly showing an association between Au-in-calcrete and biotic factors. The presence of Br (as an element commonly found in evaporite minerals) is further evidence of water being removed from this microenvironment causing precipitation. All these observations are consistent with an evapotranspiration model for the formation of Au in the calcrete; Au has been mobilised then precipitated as vadose water has been removed from the soil by trees and shrubs (Chapter 5). While the association between Au and Ca is very strong in bulk sample analyses down the soil profile, other detailed analyses on sub-samples using wet chemical, LA-ICP-MS (laser ablation inductively coupled mass spectrometry) and SXRF techniques show that it is not apparent at the sub-millimetre scale. This suggests

that the Au and Ca mobilities are similar but independent of one another and that Au does not (at the μm scale) co-precipitate with carbonate minerals.

The thesis attempted to answer a series of questions and while an attempt on these was undertaken a number of other lines of research and questions were posed that were beyond the scope of this research. Some of these are included below:

- 1) Microorganisms have been suggested to play a role in mobilising and precipitating Au in the natural environment. Recently, experimental studies have suggested that there may be a role of bacteria in the formation of Au-in-calcrete anomalies (Reith and Schmidt Mumm, 2007; Reith et al., 2009). While these studies show the theoretical involvement of bacteria there is a paucity of data describing if, how and when bacteria are involved in Au-in-calcrete anomaly formation in the natural environment. Further work in this area of research should be encouraged.
- 2) Calcium and Mg isotopes could be used to investigate the source of these elements in calcrete. Currently, Sr is used as a substitute for Ca but better detection and precision is now available to make the use of Ca and Mg more productive. Dolomite often occurs in gold-in-calcrete profiles and the mode of formation of this mineral in soils is enigmatic.
- 3) Further work on the form of gold in calcrete needs to be undertaken. The implication of this thesis was that an evaporite was playing a role e.g. Br but it is hard to believe that an organic complex is not at least playing a partial role in the formation of these anomalies given the proximity of plant roots.
- 4) All the soil profile work has been done in Australia. By sampling down a profile, particularly in early stage (Stage 1 of Gile et al., 1966) it can be clearly seen that Au and Ca are strongly related. Sampling outside Australia is suggested since we cannot discount a specific role played by Australian flora e.g. *Eucalyptus* or *Acacia* in the formation of the anomalies.

CHAPTER 8

CONCLUSION

This thesis makes several original contributions to knowledge in applied geology and specifically mineral exploration for buried Au deposits. The principal findings of this thesis and published in a series of papers are:

- 1) Gold anomalies in calcrete can form in the rooting zone of vegetation by causing the precipitation of elements due to evapotranspiration.
- 2) Gold can occur as both ionic and particulate Au in calcrete.
- 3) Gold deposition was found in the root tubules in calcareous soil.
- 4) Some Au was associated with Br in calcrete.
- 5) Gold may associate with calcrete during the life of a tree. Sand dunes containing powdery calcrete and anomalous Au can form in <26 000 years. Laminated calcrete containing Au formed in <120 000 years.
- 6) Gold-in-calcrete anomalies (with Ag) may be extended laterally due to geomorphological characteristics and create larger anomalies that may be difficult to reconcile with the original source.

Collectively these papers have shown that there are both biotic and abiotic factors involved in the formation of Au-in-calcrete anomalies which may form quickly i.e. during the age of a tree. The balance between Au-in-calcrete anomaly formation and its dilution and dispersal is in a dynamic equilibrium moderated principally by environmental factors such as climate, soil factors and geomorphology. Biotic and abiotic factors are important for mineral exploration purposes since they may serve to both generate and disperse Au anomalies in the near surface environment. While several questions concerning gold-in-calcrete have been answered, or at least addressed, several more have been posed.

In conclusion, through publication of papers, this thesis has had an impact on the scientific community and the exploration industry through greater understanding of how the process of gold in calcrete has formed and details of the parameters that affect the formation of the anomalies. Calcrete is a major exploration sampling medium in Australia and elsewhere and is regularly reported in company briefs, quarterly reports, annual reports IPOs. Companies learn about this knowledge commonly through workshops, seminars and conferences rather than the written word contained within scientific papers and theses. The impact of knowledge gained by exploration personnel

is, in my opinion, far greater from the spoken word and in-field demonstrations rather than reading the scientific literature.

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Statement

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APPENDICES

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