

# WITWATERSRAND METALLOGENESIS: THE CASE FOR (MODIFIED) SYNGENESIS

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**ABSTRACT:** The Witwatersrand (WWR) ores contain more gold than could have been derived in particulate form by erosion from any conceivable type of source area as proposed by the modified placer hypothesis. In contrast to this, syngeneses goes further to explain a host of observations from those Late Archean Au-U ores. Although recycling, placer processes, and processes of hydrothermal (diagenetic/authigenic) mobilization all contributed, syngeneses was a major factor contributing to ore genesis in this huge metallogenic province. Over 80% of the gold occurs in the Main Reef and Bird Reef of the Johannesburg Subgroup in the Central Rand Group, and about half of this gold is closely associated with carbon derived from microbial remains. In the principal deposits within the WWR basin, the ore is disposed in thin carbonaceous horizons of extensive lateral continuity upon chronostratigraphic unconformities in otherwise unmineralized siliciclastic metasediments. The ore-bearing horizons are not themselves part of the erosion cycle that gave rise to those paleosurfaces but were generated during the initial phase of renewed cycles of deposition after long intervals of nondeposition. They bear little resemblance to placers, their alluvial character seemingly inherited from reworking in fluvial environments.

Most of the gold and probably also part of the uranium were made available for transport in solution under relatively low-temperature, chemically aggressive environmental conditions, a situation favored on the emerging Kaapvaal Craton. Intense chemical weathering was made possible by the influence of the same ionizable gases as occur in geothermal systems, and this was a crucial factor leading to metallization. These elements, together with a host of other heavy metals, were then transported to the edge of the depository. A key confluence of conditions was completed with the blooming of microbial communities during hiatuses in sedimentation. Over large areas, microbial mats developed directly on paleosurfaces upon which the goldfields occupy slight depressions, bounded on either side by clean quartz arenites. The resulting metallization was a complex chemical and biochemical precipitation of gold, uranium, pyrite, and associated Co, Ni, Cu, Pb, and As in thin, areally extensive deposits. Metallization was focused at several carbonaceous horizons along the north and northwestern margins of the WWR basin, depending on the availability of metal-rich aqueous fluids coincident with the stillstand of land surface degradation and the consequent proliferation of microbial mats. Biochemical processes supplemented low-temperature geochemistry of the fluids in helping to concentrate a substantial portion of WWR gold in larger particles, which were transported further downslope and then subjected locally to fluvial processes. Gold precipitated outside of the preserved basin by these processes likewise will have undergone alluvial reworking prior to deposition in the conglomerates without the originally associated carbon; recognition of this feature diminishes the source rock problem. Minor remobilization of metals occurred during diagenesis and metamorphism.

**KEY WORDS:** syngeneses, metallogenesis, Witwatersrand, gold, uranium, lithofacies, biogeochemical processes

“...on the one hand, against the idea of a simple placer-deposit, we must set the fact that none of the pebbles, however big, ever contain gold: on the other, the theory of posterior impregnation does not tally with the constant presence of rolled pyrites. Thus we are driven back on the second hypothesis, that is, chemical precipitation of the gold and pyrites during the actual process of sedimentation.”—De Launay (1896, p. 83)

## INTRODUCTION

In an early and remarkably perceptive work, De Launay (1896, p. 79) ruled on the question of whether Witwatersrand (WWR) gold “...was formed either before, at the same time as, or else after the conglomerate.” Paraphrasing this, the popular options for origin

remain: paleoplacer, synsedimentary, and hydrothermal. Historically, the first and third theories have received far more attention than the second. Thus, Frimmel and Minter (2002, p. 38) and Frimmel et al. (2005a) succinctly contrast arguments in support of the paleoplacer and hydrothermal hypotheses but fail to acknowledge any contribution whatever by syngenetic processes. Accepting the term “syngeneses” (Chilingar et al. 1967, p. 322) for the “...processes by which sedimentary rock components are formed simultaneously and penecontemporaneously...,” we argue here for a dominant role for syngeneses: Specifically, the strata constituting the WWR Supergroup are the products of an evolving crust, and the metalliferous deposits formed in response to certain stages in development of the atmosphere, hydrosphere, and biosphere.

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Overviews of the WWR goldfields and their geological setting are matters of comprehensive record (e.g., Pretorius 1981a, Tankard et al. 1982, Robb and Meyer 1995, Handley 2004, Frimmel et al. 2005a). The Witwatersrand Basin formed between 2.97 and 2.714 Ga (Minter 2006). Frimmel and (Minter 2002, p. 106) summed up evidence for the alluvial structure of the ore and argued cogently for the modified placer model. That qualification recognizes the pre-WWR origin of the gold and that some of the gold particles are unequivocally of detrital origin (Hallbauer and Utter 1977, Minter et al. 1993, Minter 1999). In confirmation of the latter inference, three-dimensional (3-D) X-ray photographs of various reef samples reveal gold grains that closely follow all sedimentological features of a placer (Hallbauer and Barton 1987). There are also clear indications that, at least locally, gold has undergone some hydrothermal mobilization (Ramdohr 1958, Frimmel and Minter 2002). Thus, ore genesis cannot be construed as the result of any one single process. Furthermore, numerous deposits of "Witwatersrand type" of various ages are known (Mossman and Harron 1983, Falconer et al. 2006, Minter 2006, Reith et al. 2006), although all pale by comparison with the scale of the "real thing." Simply stated, the Late Archean WWR sedimentary basin is a huge ore province of ~30,000 km<sup>2</sup> (Figs. 1, 2) in which certain ore-forming processes worked extraordinarily efficiently. The question is: What exactly were those processes?

Paradoxically, throughout the basin, the pebbles and quartz arenite, which constitute 95% to 97% of the common quartz-pebble reef, are explained as debris resulting from erosion of gneiss-granite-greenstone terrain, whereas the other 3% to 5%, composed of pyrite and a complex economically important suite of minerals in the reef matrix, remain enigmatic. According to Hallbauer and Utter (1977), detrital gold particles liberated from the conglomerates show signs of transport not

exceeding 35 to 40 km. Gold and uraninite occur with sulfides in the conglomerate matrix and not in quartz, neither in quartz pebbles nor in original sand grains. Except on the scale of hand samples, there are no occurrences of gold only, or of uraninite only.

From the inception of mining to 2002, the WWR has produced a total of 49,332 metric tonnes (t) Au at an average recovery grade of ~8 g/t (Handley 2004). Individual reefs contain up to 1200 g/t. The gold occurs in several distinct morphologies as discrete particles and commonly forms some intergrowths on (and rarely in) pyrite; the average size of gold particles is about 120 μm (Hallbauer and Barton 1987). Fineness ranges from 650 to 960. Hg content can reach several percent (Frimmel et al. 2005a). Uranium, which has on average a recovery grade of ~271 ppm (Frimmel et al. 2005a), occurs as uraninite and brannerite. Pyrite, the most common sulfide among a variety of sulfides of As, Co, Ni, and Pb (and less commonly Cu, Zn, and Mo), occurs in several forms, and virtually all of them are auriferous, though none more so than a rounded dull type of so-called "buckshot." Friable, banded, and porous, this pyrite has all the characteristics of a chemical precipitate, including trace amounts of a range of metals present as major constituents in accompanying sulfide minerals (Feather and Koen 1975, Hallbauer 1986). The sedimentary control and remarkable stratigraphic continuity of this complex ore assemblage, in common association with carbon and the sheer mass of gold found, led to the alternative source concept that the key components gold and partly also uranium were derived from the hinterland, carried in solution to the edge of the basin, and precipitated there under reducing conditions (e.g., Sharpe 1949; Koen 1961; Reimer 1975, 1984; Simpson and Bowles 1977; Mossman and Dyer 1985; Horscroft 1989, 2004). Note that in this scenario, any gold precipitated outside of the preserved basin could have been reworked

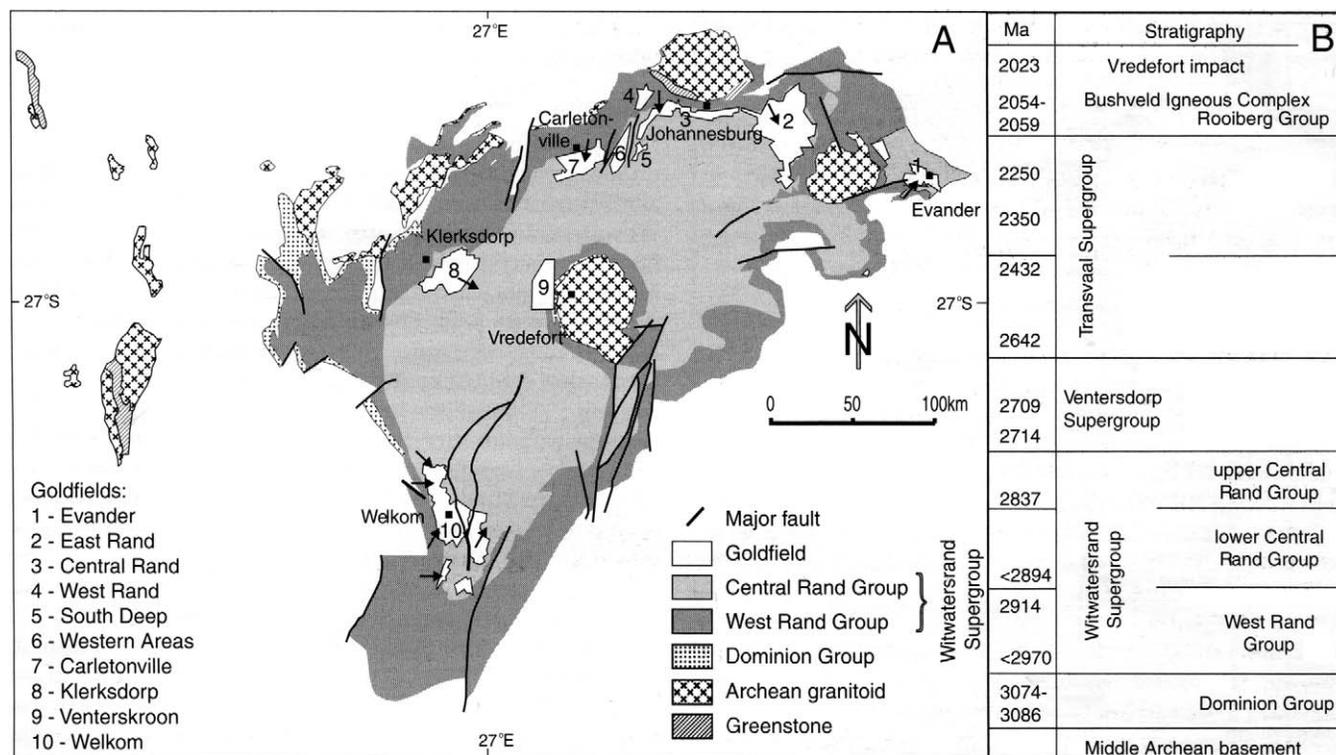


FIG. 1.—Simplified surface and subsurface geological map of the WWR basin showing location of the main goldfields, major faults, and the main stratigraphic units and ages (see Frimmel and Minter [2002] for references). Arrows indicate paleocurrent directions of reefs in the Central Rand Group (modified after Frimmel and Minter 2002).

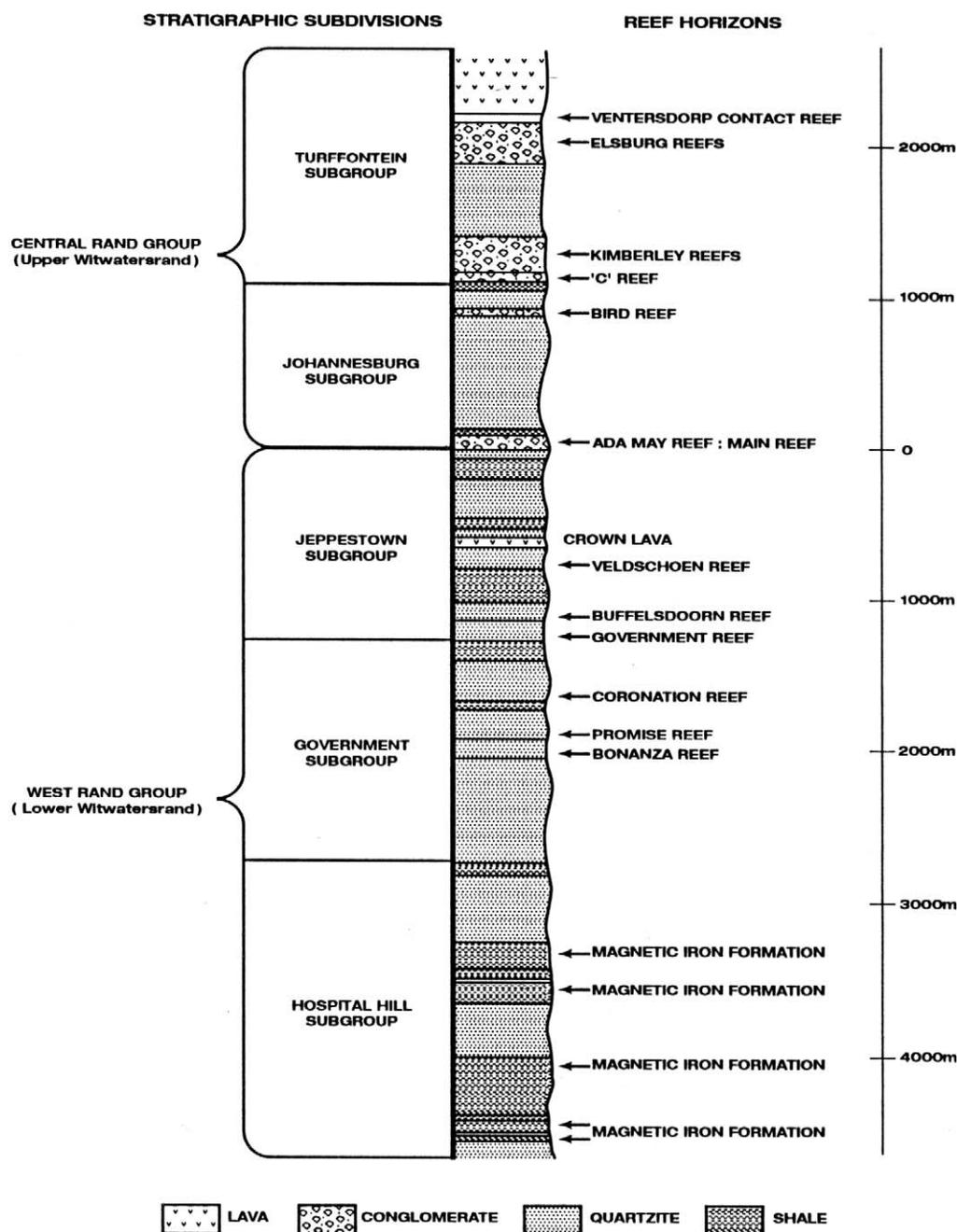


FIG. 2.—Generalized stratigraphic section of the WWR Supergroup (based on the Klerksdorp District) contrasts the extent of the West Rand Group and the Central Rand Group, showing the numerous reefs, including the Bird Reef (equivalent to Vaal Reef and Basal/Stein Reef) and the Main Reef (equivalent to the Carbon Leader) of the Johannesburg Subgroup, which together account for >84% of Witwatersrand gold production. The Bird Amygdaloid (not shown) occurs as a thin layer above the Bird Reef. (After Horscroft 1989.)

into detrital grains and deposited by normal processes within the basin and without the originally associated carbon; in this instance, the Au/Ag ratio, for example, will have been strongly influenced by the *locus precipitandi*, which might well lie outside of the preserved basin. Redistribution further into the basin inevitably followed locally by normal fluvial processes.

Broadly viewed, three environments of deposition occur throughout the West Rand Group (WRG) and Central Rand Group (CRG) of

the WWR Supergroup (Figs. 1, 2): (1) reefs in the WRG, which have characteristics resembling reefs of the underlying Dominion Group and of the Pongola Supergroup; (2) carbon reefs along the base of the CRG (i.e., the Main Reef horizon), and remarkably similar reefs at the higher Bird Reef horizon, which together have produced the bulk of the gold to date, and the horizons of which can be traced through the greater part of the northern and western portions of the basin; and (3) upper reefs of the CRG, particularly the Eldorado/Elsburg, which also

contain some very large deposits. We perceive a common genetic theme in all these environments. Here, we focus on the case for syngensis of much of the WWR gold, the objective being to demonstrate that the case for this genetic model is more fully compatible with geological and mineralogical observations than either of the two competing models.

### THE MESO- TO NEOARCHEAN ENVIRONMENT

Current models of Precambrian atmospheric evolution, though differing in detail, all consider the importance of methane (e.g., Tice and Lowe 2004, Kasting 2005). Ueno et al. (2006) documented  $^{13}\text{C}$ -depleted methane in hydrothermal inclusions in ca. 3.5 Ga cherts from the Pilbara Craton, Australia. It is therefore reasoned that prior to 3.8 Ga, global deep freeze resulting from the “faint sun” would have been avoided by a  $\text{CH}_4$ - and  $\text{CO}_2$ -rich greenhouse atmosphere due to volcanic outgassing and minimal drawdown resulting from weathering. Kasting (2005) argues that on a colder Earth, silicate weathering would have been inhibited, as would aqueous transport and sedimentation, whereas volcanism would have continued. Reduced gases such as  $\text{CO}$ ,  $\text{CH}_4$ , and  $\text{H}_2\text{S}$  would have been oxidized by photolysis of water vapor to the hydroxyl radical. Anaerobic bacteria, probably one of the first life forms on Earth, appeared after 3.8 Ga, resulting in increased surface temperatures (Schidlowski 2001, Kasting and Siefert 2002, Kasting 2005), and promoting a positive feedback loop and increased bacteria populations. In turn, increased temperatures would have led to increased weathering and decrease of  $\text{CO}_2$ , an important feature consequent to the emergence of a major continental landmass ca. 3.1 Ga, of which the Kaapvaal Craton was part. Then, according to Lindsay (2008, p. 836), expansion of the biosphere shortly after 3 Ga locally led to at least low levels of oxygen for the formation of banded iron formations (BIFs), and allowed the formation of humic acids.

In his comprehensive review of evidence bearing on the Meso- to Neoproterozoic (3.1 to 2.6 Ga) atmospheric evolution, Frimmel (2005) highlighted the WWR basin on the Kaapvaal Craton as providing clear evidence of an acid and reactive anoxic atmosphere enriched in  $\text{CO}_2$  and  $\text{CH}_4$ . Otherwise conducive to intense chemical weathering, this situation supposedly allowed survival of detrital uraninite and pyrite, neither of which has traditionally been considered to be capable of surviving exposure under oxidizing atmospheric conditions. This is, however, a false presumption, given that Tertiary quartz-pebble conglomerates in southern New Zealand's Belle Brook deposit (Falconer et al. 2006) contain abundant detrital sulfide minerals, some of which (specifically pyrite and arsenopyrite) are of “long-distance” origin (Falconer et al. 2006, p. 525), together with diverse examples of look-alike WWR-type grains of gold. Uranium here is also present, to 17 ppm, in close association with diagenetic marcasite (D. Falconer, written communication, April 2010). Further, Falconer et al. (2006) report that diagenetic sulfides (and the gold) from the Belle Brook quartz-pebble conglomerate are compositionally similar to those of the WWR; concretionary textures (mostly of marcasite) of the Belle Brook sulfides and the gold “forms” likewise strongly resemble WWR samples (ibid, p. 541).

In WWR ores, according to Minter (1990), uraninite is best preserved in kerogen, though in the matrix of auriferous conglomerates, coatings of gersdorffite or titania outline allogenic uraninites. He also observed that onlapping unconformities (in the Welkom Goldfield) are barren of uraninite, which was destroyed by weathering. Thus, despite a reducing atmosphere, Earth's surface, at least locally, was exposed to oxidation processes. One such process is bacterially mediated, which, in taking iron from the ferrous to the ferric state, oxidizes  $\text{UO}_2$  (Wadden and Gallant 1985, p. 129).

Various workers (e.g., Wiebols 1955, Minter and Loen 1991, Rust 1994, Young et al. 1998) have proposed that glacial conditions occurred during WWR times. Evidence of glaciation is preserved in

two tilloids in the Government Subgroup that are widely correlatable across the basin. Striated pebbles and boulders to 50 cm diameter are common, and a borehole intersection of a 3-m-diameter boulder is recorded; carbonate layers are also present (Antrobus, unpublished data, 1980). However, bearing in mind Kasting's (2005) observation (stated previously herein) and considering that weather conditions must have been such as to allow for the copious growth of microbial mats, the intensity of that glaciation is open to question.

In the WRG, nonsulfidic magnetic shale with appreciable carbonate occurs at intervals. Examples include the Contorted Bed, and the Coronation Formation and Promise Formation in the Government Subgroup, confirming the presence of dissolved iron in seawater. That seawater will also have contained gold, probably in amounts exceeding modern conservative estimates, ranging from average concentrations of 5 to 50 ppt (parts per trillion). Altogether, BIF material is estimated (Reimer, unpublished data, 2009) to comprise 10% to 15% of the lower part of the WWR succession, and it will have served as a sink for some of the early production of oxygen by photosynthesizing microbes. In contrast, sulfides, and not oxides, occur chiefly in Algoman-type iron deposits, a feature attributable to their local hydrothermal derivation or sediment-hosted sulfate reduction (Canfield 2005). According to Nisbet and Sleep (2001), important early energy sources arising in both oceanic and terrestrial hydrothermal systems included  $\text{H}_2$ ,  $\text{H}_2\text{S}$ , and native sulfur. High heat flow was characteristic of Meso- to Neoproterozoic time, and among the mix of volcanic gases,  $\text{H}_2\text{S}$  driven from hydrothermal emanations would have been readily accessed by sulfide-oxidizing anoxygenic phototrophs and/or facultatively aerobic microbes in surficial as in subsurface terrestrial settings. Sulfur isotopic compositions of pyrite ought to reflect this situation. Their wide range (between  $-5.3\text{‰}$  and  $+6.7\text{‰}$ ), as reported by England et al. (2002) and Canfield et al. (2000), actually lies within the range of typical Archean sulfide deposits. While this is not surprising, the presence of mass independent fractionation, according to Bekker et al. (2004), suggests that atmospheric oxygen was not present at significant levels. Phillips and Law (2000) attributed the  $^{34}\text{S}$  heterogeneity to diagenetic mixing of meteoric (sulfate) and hydrothermal (sulfide) fluid phases, but bacterial sulfate reduction could have achieved similar results. In the CRG, carbonate occurs as dolomitic shale and calcareous quartzite in the Booyens Shale (Antrobus et al. 1986), and gentle synclines in the floor of the Main Reef Group at Durban Roodepoort Deep mine contain dolomite (Pretorius 1964). The great thickness of arenites in the CRG has no iron minerals, except for pyrite, which is confined to thin mineralized horizons correlatable across the basin as chronostratigraphic horizons.

Facultative aerobes were present during WWR sedimentation and earlier (Noffke et al. 2006a, 2006b; Mossman et al. 2008; Noffke 2008). In addition to incontrovertible evidence in the form of stromatolites (Walter et al. 1980), and fossil biofilms in the Nhlazatse Section (2.9 Ga Pongola Supergroup) relict from early Mesoarchean mat communities (Noffke 2008), molecular evidence now confirms that by the Late Archean, archaea occupied the subsurface biosphere (Ward et al. 2004). Specifically, archaeal- and bacterial-related lipid carbon skeletons (including diasteranes, tricyclic terpanes, steranes, and hopanes) have been found in kerogen from the 2.7-Ga-old metasedimentary rocks of the Porcupine Gold Camp in Timmins, Ontario (Ventura et al. 2007). Minor nonphototrophic oxygen sources could have included  $\text{CH}_4$  photolysis (Catling et al. 2001) and disproportionation of atmospherically derived  $\text{H}_2\text{O}_2$  by early catalase enzymes (Canfield 2005, p. 3-6). Consequently evolution of the Meso- to Neoproterozoic atmosphere will have been extensively modified by microbial activity. Indications are that microbes also played key roles in ore genesis (Mossman and Dyer 1985; Hallbauer 1986; Mossman et al. 1999; Reith et al. 2006, 2007). The effect of the biosphere—both surface and subsurface—is highlighted by various important ore-

bearing biomarkers and the preservation of oil-bearing fluid inclusions in WWR ores (Dutkiewicz et al. 1998, Mossman et al. 2008).

The repeated deposition of a complex suite of ore minerals within extensive vertical and lateral intervals reflects a regional control of ore-forming processes. Unlike many carbonaceous ores, particularly those associated with volcanic rocks and BIFs, WWR ore is nonrefractory (to cyanide extraction), and the similarity in mineralogy permits the application of common metallurgical recovery processes throughout the five goldfields. Volcanic degassing during WWR time would have led to accelerated chemical weathering and the production of ionizable acid gases such as those that are common to geothermal systems (i.e., CO, SO<sub>2</sub>, H<sub>2</sub>S, CO<sub>2</sub>, HCl, HF, H<sub>2</sub>, and CH<sub>4</sub>). This is important because these gases would have formed solutions of hydrothermal composition similar to those that generate epigenetic multimetal deposits complete with mineral zonation (a feature conspicuously absent in WWR ore). We hold that this mechanism was one of the major factors contributing to WWR metallization (Fig. 3). Certainly, of all ingredients, sulfur would not have been in short supply. Core logs (Antrobus et al. 1986) reveal the occurrence of hundreds of meters thickness of argillaceous rocks (containing chlorite, chloritoid, pyrophyllite, K-mica), including some intervals of magnetic mudstone, within the sterile siliciclastic metasediments south of the Central Rand. However, within the latter, the markedly pyritic auriferous ores survived; they lie preferentially along chronostratigraphic horizons focused mainly in the Main Reef and the Bird Reef. Broadly viewed, the ore is the product of the combined effects of atmosphere, biosphere, and hydrosphere.

Regardless of the tectonic setting (intracratonic basin on a stable craton, or foreland–back-arc environment), the twofold dominance of WWR gold production (Fig. 4) over that of greenstone belts worldwide is of outstanding interest and commands consideration (Pretorius 1981b, Woodall 1988).

### CRG LITHOFACIES

Extensive quartzitic deposits are the main Archean sedimentary rocks overlying the Kaapvaal Craton. Formed under extreme weathering conditions, only the most durable and chemically inert material is preserved in the WWR arenites and conglomerates. Consequently, black sands are absent. In brief, two factors are of paramount importance (Reimer and Mossman 1990, p. 426): “First, recycling of older sedimentary material was critical to the genesis of the conglomerates; about 60% of the source area consisted of arenaceous sequences. Iron-titanium mineral grains from this source had been altered to rutile-leucoxene prior to erosion, and thus did not contribute fresh iron-titanium minerals to the conglomerates. Second, those minerals derived from the remaining 40% of the source area were altered and decomposed to rutile-leucoxene in the Witwatersrand conglomerates. Furthermore, much of the resulting finely dispersed material helped to form brannerite, an important titanium sink.” An important corollary: Sulfidation of black sands does not account for WWR pyrite and the lack of detrital Fe-Ti-oxide minerals.

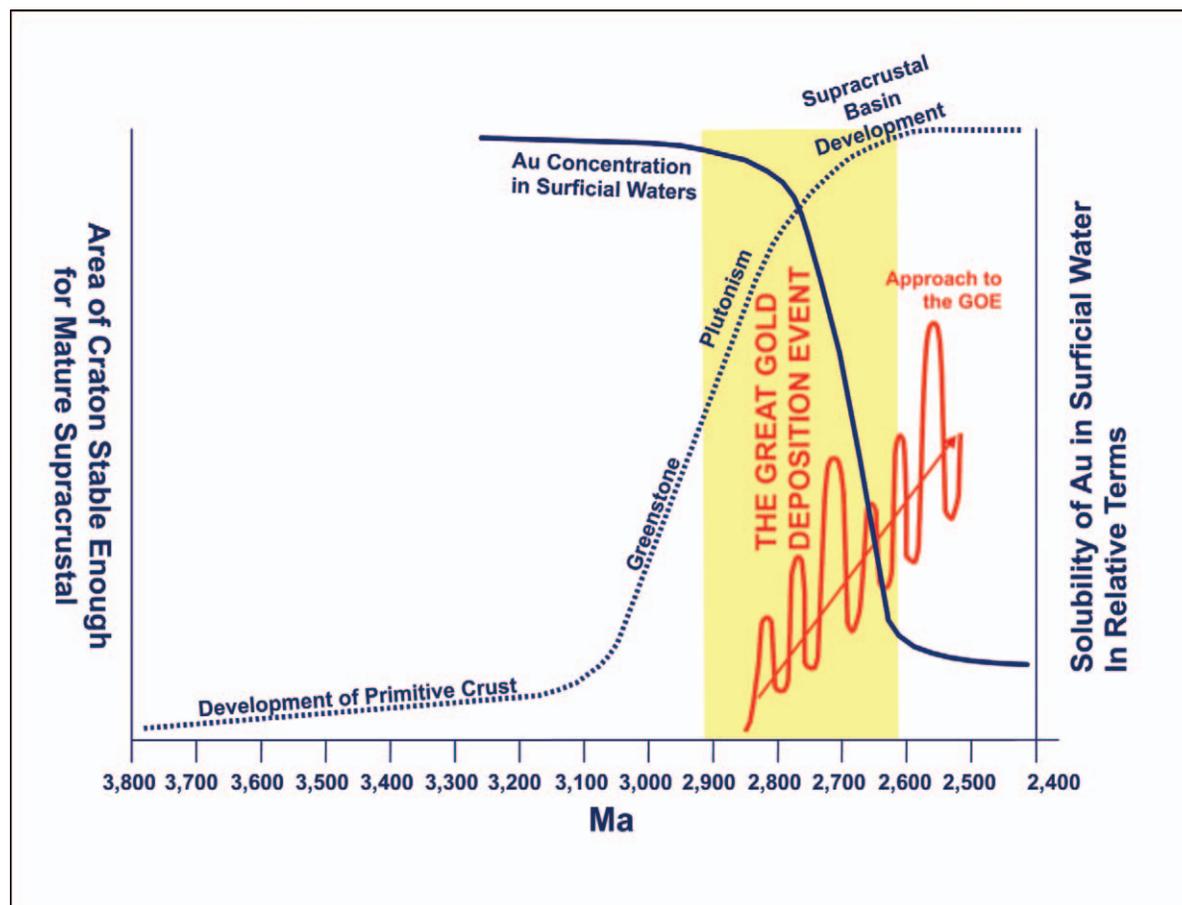


FIG. 3.—Cartoon outlines circumstances leading up to (and overlapping) “The Great Gold Deposition Event,” which was the gold equivalent of the peak development of BIF during the “Great Oxidation Event.”

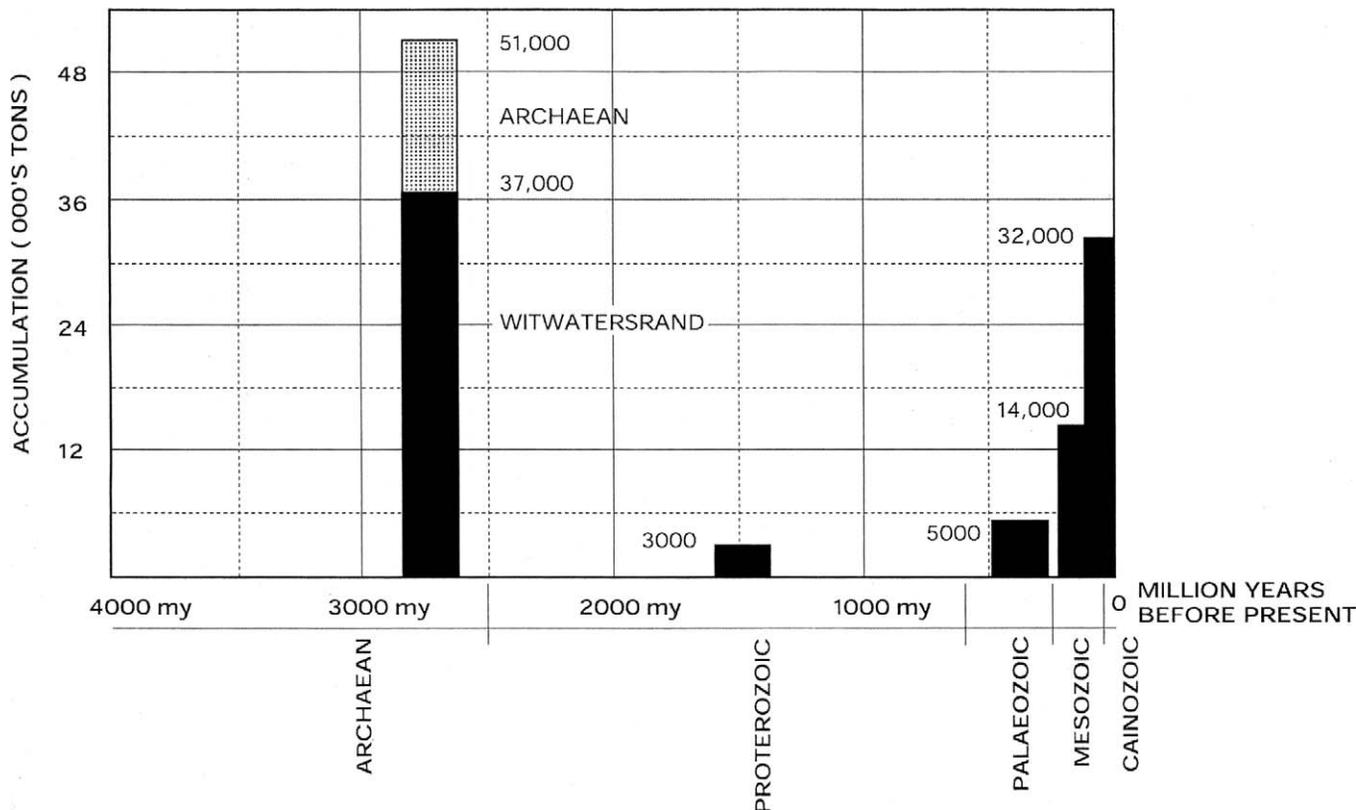


FIG. 4.—Proportion of gold production from Archean deposits in this graph (“Gold in Geological Time”) shows an approximately twofold production of the Witwatersrand goldfields over that of greenstone belts worldwide. (After Woodall 1988.)

In the conglomeratic ores, the small range of pebble types, dominated by quartz and chert (locally with lesser porphyry), is reflected also in the small range of detrital minerals, notably sulfides, zircon, chromite, tourmaline, and platinum group minerals (PGM). Pebble size too is a significant aspect; closely packed and even-sized pebbles are characteristic of many reefs. For example, the Carbon Leader Reef in the West Wits Line (Carletonville) Goldfield rarely exceeds 5 cm in thickness and exhibits little variation in pebble size (mean diameter = 1.13 cm); this may be a possible consequence of milling under steady winds in shallow water. (Note: Old names of Witwatersrand goldfields and gold mines are given in parentheses.)

In general, all WWR ore bodies are composed of mature pebbly arenite comprising lithofacies ranging from clast-supported oligomictic (far fewer polymictic) conglomerate to loosely packed matrix-supported conglomerate, pebbly arenite, or pebble lag surfaces associated with cross-bedded quartz arenite (Frimmel and Minter 2002, p. 23). As described in the following subsections, there are overall essentially three sedimentary facies of similar metal content, but of markedly different physical character.

#### *Quartz-Pebble Reef (Conglomerate) Facies*

According to Pretorius (1975, p. 45), the quartz-pebble reef facies is the result of a three-stage cycle of sedimentation—progradation, aggradation, and degradation—on an alluvial fan. Open-work gravel deposited during the first pulse (regression) was succeeded by the infiltration of heavy minerals carried in the sand fraction of alluvium (transgression); end of cycle (stillstand) washing concentrated residual heavy minerals on erosion surfaces. In consequence, much of the

primary mineralized sediment was not preserved and is restricted to relatively small quartzite bodies and to quartzite phases of the quartz-pebble reef. Fluvial reworking concentrated the heavy minerals as lag gravels and the basal portions of reefs were commonly scoured surfaces; both features have been mistaken to be proof of detrital (placer) origin. Washing of the fluvial sand led to infiltration of the heavy minerals between the pebbles, resulting in widely mineralized auriferous quartz-pebble reef, giving the name to this class of ore (see Fig. 5A–C). This process is widely accepted. It is described in detail by Smith and Minter (1980, p. 7) in the Elsburg No. 5 Reef in the upper section of the Central Rand Group and was applied by Tainton and Meyer (1990) to the “small pebble” Promise Reef in the middle section of the West Rand Group. The highest gold concentration in the pebble reef occurs in well-washed, well-sorted, and clast-supported pebble layers. Reefs range from single conglomerate to composite conglomerate with interbedded quartz arenite.

Some reefs are locally reduced to a single line of small pebbles, suggestive of formation as a pebble carpet formed by sustained eolian activity in flat fluvial (braided) streams and in shallow lagoonal environments. Such environments were considered by Pretorius (1975) to have favored growth of algal mats as generally accepted by sedimentologists. Not surprisingly, ventifacts (dreikanter), including toroidal gold particles, have been identified (Minter et al. 1993, Minter 1999) in cross-bedded quartzite of the Basal Reef, and they are held as unequivocal evidence of the detrital nature of at least a portion of the gold. These toroids have not necessarily undergone long transport. Consider, for example, the case of auriferous muddy froth blown ashore from the sinter flat of New Zealand’s Rotokawa geothermal system. According to Krupp and Seward (1987, their Fig. 13, p. 1127),

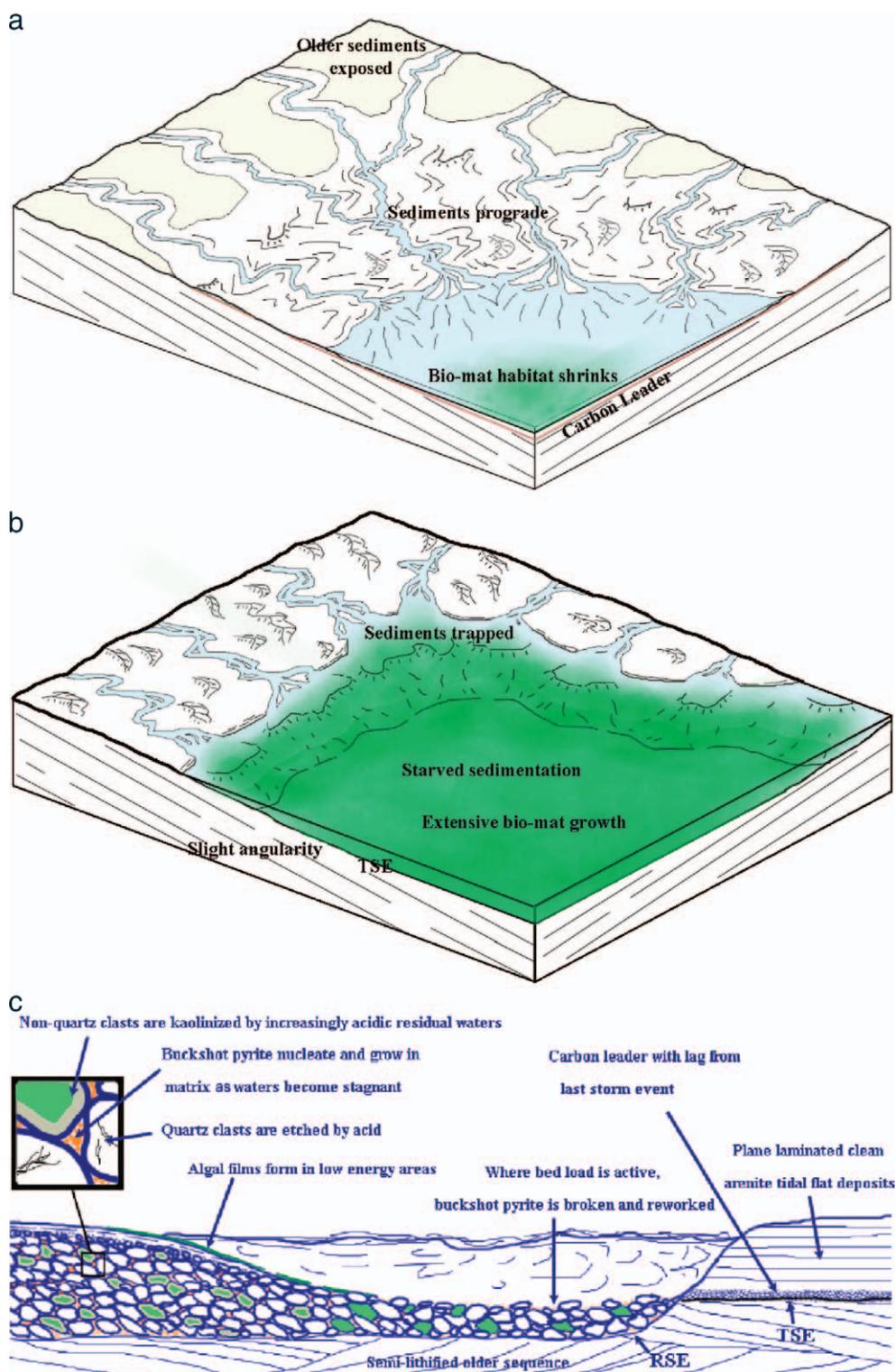


FIG. 5.—Cartoons illustrate: (A) regressive and (B) transgressive cycles of sedimentation, and the response of biomat communities to these processes, and (C) examples of developments, as indicated, upon successive transgressive surfaces of erosion (TSE) and regressive surfaces of erosion (RSE) on a WWR alluvial fan.

the surface film forming the froth assays up to 50 ppm Au. By this process, small flakes of gold might easily be transformed through eolian deflation from crustified auriferous biofilm (whatever its origin) into toroids.

In the Central Rand Group, the most important economic horizons extend laterally up to 80 km. In the West Wits Line (Carletonville) Goldfield, the Carbon Leader Reef extends through 15 km strike. From the West Rand eastward through the Central Rand and the East Rand, the Main Reef comprises three horizons, which merge eastward by transgression of the uppermost to a single (south) reef. Each of these horizons was reported to have stretches of thin carbon reef with high gold content, but high grade sectors were also recorded in close-packed, well-sorted, quartz-pebble reef.

An exception to Pretorius' three-stage cycle of sedimentation might be the thick ore zone(s) in the Elsberg Formation, which developed in a reworked placer on the Eldorado paleosurface near the hinge of several unconformities in the Free State Area (Welkom) Goldfield (Minter et al. 1988, p. 488). However, this zone consists of numerous thin, closely spaced reefs. The Eldorado paleosurface records the last major erosion interval. In this instance, Minter et al. (1988) show that all three types

of pyrite (round compact, round porous, and concretionary or layered) are detritally rounded, as are grains of broken columnar kerogen. The results of detailed mineralogical work by Tucker (1983) on this Eldorado ore support the intimate association of gold (and carbon) with syngenetic "buckshot" pyrite (Fig. 6).

Overall, the morphology of the principal deposits is remarkably similar (Minter et al. 1986, Minter and Loen 1991). A typical feature is the occurrence of one principal reef on a braid delta, commonly with lesser associated reefs as lobes, and coalescing lobes of "placer" sediment up to 30 km wide, merging with shoreline environments. Lobes occur at different stratigraphic levels, as debris flows and terraced fluvial deposits (Liebenberg 1955). In some environments, several lobes occur on the same paleosurface. Typically, lobes may adjoin, overlapping slightly with some erosion of older by younger, though they can have divergent sediment-transport directions. In the East Rand Goldfield, and also in the Free State Area (Welkom) Goldfield, some reefs cannibalize (i.e., overlie and cut across) older reefs (Minter and Loen 1991, p. 77-78). Pebble composition too can vary, some oligomictic, others polymictic, but all are dominantly made

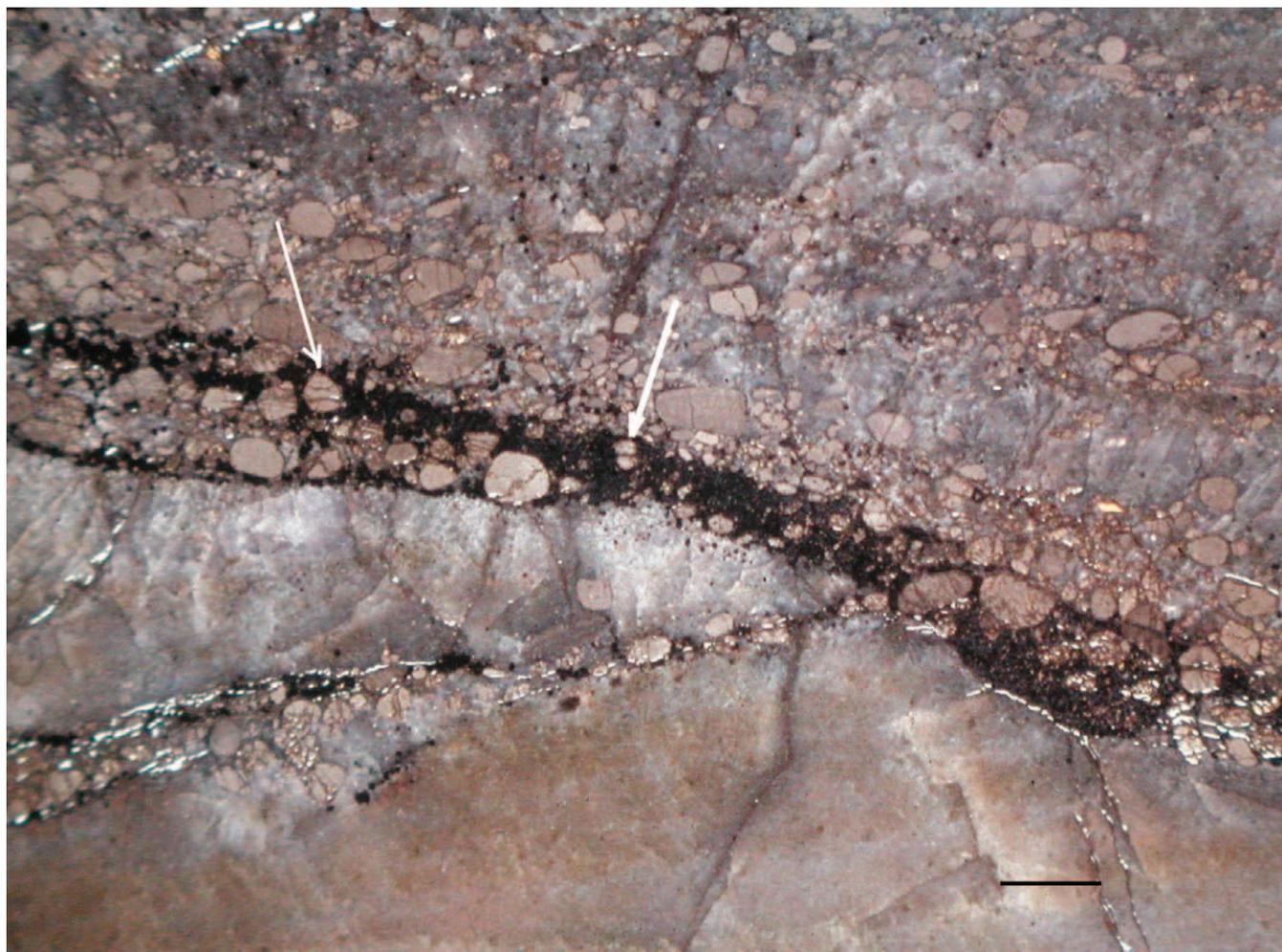


FIG. 6.—Lower portion of the Elsberg Reef, Cook Section, Randfontein Estates Mine, showing rounded detrital pyrite (arrows) floating in a thin carbon seam. This is one of many common, but subtly powerful, syngenic features illustrating their common sedimentary origin. The carbon contains filamentous gold. Note that some of the pyrite pebbles show imbricate structure (flow from left to right). Wedging and truncation of arenite layers (left-hand side) may be due to cross-bedding. Scale bar = 1 cm. (Photograph by D.K. Hallbauer.)

up of quartz and chert. Throughout the deposits, the ore minerals are similar, although proportions may change.

### *Quartzite (Quartz Arenite) Facies*

This ore facies was mined as discrete small deposits in the East Rand Goldfield (De Jager 1986, p. 125) and the Free State Area (Welkom) Goldfield. In the Steyn placer of the latter goldfield, quartzite contains about 11% of total Au and five to ten times more U than Au. The lower parts of horizontally laminated quartz arenite are characterized by numerous pyritic laminae and sporadic carbon laminae, both of which are enriched in Au and U. Similar sediments form a common and laterally extensive facies within many of the channel placers. According to Minter et al. (1986, p. 531), from 5% to 25% of total mineralization is concentrated in the quartzite facies of the Carbon Leader Reef.

In many quartz-pebble reefs, gold is concentrated in the upper (top) portion of the reef, illustrating Pretorius' (1975) perception that the gold came in with the sand - an observation first recorded by Pirow (1920), referred to by Frimmel and Minter (2002, p. 531), and which is widely accepted by placerists. A critical aspect is that the quartz bodies have the same overall composition as the quartz-pebble reefs so that the answer to the question of genesis can be said to be "written in the sand."

Top concentration is also common in Jacobina, Bahia State, (Brazil), where ore has been dated at 2.86 Ga—in a concordia plot of four samples from the Main Reef zone—by H. Welke (unpublished data, 1986, Bernard Price Institute, Johannesburg). The ore is mineralogically similar to that of the WWR, although carbon only occurs as sparse "flyspeck" grains. The host rock is a clean recrystallized quartz arenite, similar in composition to that in the WWR, likewise without black sands.

### *Carbon Facies*

The most highly mineralized reefs are those enriched in carbon (Handley 2004). They provided the infrastructure for the gold mining industry. As summed up by Gregory (1962, In Greathead and Van Roggen, 1986, p.235): "The Basal Reef is by no means the only gold-bearing reef discovered, but it is the only generally payable reef, upon which the entire edifice of Free State mining, and, with it, the impulsion which mining has given to the Free State generally, now rests."

These are remarkable ore-bearing horizons, although they bear little if any resemblance to placers. The Main Reef horizon extends for a distance of about 80 km, with a thin seam of carbon only a few centimeters thick, along several sectors. This seam is overlain by sand (quartz arenite) and small pebbles about 2 cm in diameter, and it carries hundreds of grams of gold per tonne and considerably more uranium over a thickness of 5 cm. At intervals along strike, the seam has been eroded and replaced by layers of similar small pebbles, and a 30 cm thickness of quartz sand in shallow channels. Mossman et al. (2008) showed that the so-called "carbon" is commonly a mixture of kerogen (fossil organic matter) and derivative (now solid) bitumen, and is thus of indigenous origin. A direct genetic association via biomineralization between the gold and the carbon has been postulated by various workers (Mossman and Dyer 1985, Hallbauer 1986, Reith et al. 2007, Mossman et al. 2008).

The high concentration of gold and uranium in the carbon facies is illustrated by the analysis of a sample of the Carbon Leader Reef, Western Deep Levels, West Wits Line (Carletonville) Goldfield, which reports: 1250 ppm Au; 58,500 ppm U; 4240 ppm Th; 250 ppm Ag; 57.05% C; U/Au = 47/1, and U/Th = 14/1 (O'Connor and van Zyl 1985). Frimmel et al. (2005a, p. 3) noted that ore from Carletonville Goldfield, at an average grade of 31 g/t over its history, is the richest of

any WWR goldfield. Extraction of the characteristically thin WWR ore introduces large amounts of waste, which may exceed the volume of ore by up to 25 times (or more), with the result that recovered grade greatly understates the in situ grade.

A key feature of many reefs, and carbon reefs in particular, is their relation to preexisting topography. Over large areas, carbon seams occur directly upon paleosurfaces (unconformities, disconformities, shallow erosion channels), with the goldfields occupying slight depressions of those surfaces. The deposits are not linked to degradation (which could be construed as placer forming) that formed the paleosurfaces but are associated with carbon seams formed directly upon those paleosurfaces. Gently inclined paleosurfaces controlled the prevalence of braided stream environments and the conditions that led to the accumulation of unusually continuous, well-sorted, well-mineralized, quartz-pebble reefs (Fig. 7A, B). The depressions are essentially wide, shallow valleys eroded into footwall sediments. Angular unconformities are typically barely discernible; for example, the gradient of the paleosurface beneath the Vaal placer in the Klerksdorp Area (Klerksdorp) Goldfield changes downdip over 20 km from only 0.010% to 0.003% (Tankard et al. 1982, p. 134). In some instances, a pencil line-thick layer of gold accompanies carbon along the contact with the paleosurface (see Fig. 8A, B).

A thin quartz arenite is present in the hanging wall of each of the principal carbon reefs: [e.g., the Main Reef Leader/South Reef of the Central and East Rand, the Carbon Leader at Carletonville, the Vaal Reef at Klerksdorp, and the Basal Reef (Bird horizon) at Welkom]. This unit is referred to variously as "siliceous quartzite," "orthoquartzite," or "placer quartzite". At the Doornfontein Mine (Carletonville Goldfield), where the quartz arenite is 2 m thick, Engelbrecht et al. (1986, p. 625–627) describe it as "...a clean, massive, medium- to coarse-grained, siliceous quartzite, completely free of cross-bedding, argillaceous [*sic*] partings, and bedding planes." It is the product of extreme weathering in proximal source areas, producing pure, clean sands that, with progradation of the basin, came to rest directly over the reef and constitute the hanging wall. The reef formed on a clearly defined paleosurface of underlying quartz arenite, with a 1- to 5-cm-thick carbon seam along the contact and, in places, a film of gold, 1 to 2 mm thick, on top of the carbon. There is a widespread association of the carbon reef with small quartz pebbles, 1.5 to 2 cm in diameter, which rest directly upon the paleosurface in some areas. At 2 to 4 cm in diameter, the pebble size in the quartz-pebble reefs is larger than that in the carbon reefs. A prolonged interval of extreme weathering during generation of the hanging-wall quartz arenite would have allowed ample time for the contemporaneous flourishing of microbial mats upon the paleosurface, an event which played a critical role in the genesis of the extremely rich ore along this horizon. The features described here are small-scale aspects of the larger WWR, where highly mineralized thin reefs are interspersed between quartz arenites of such purity that there is scarcely any indication of the nature of the source rocks.

## METALLIZATION

### *Mineralogy and Geochemistry*

Concerning various pre-WWR pyritic quartz-pebble conglomerates in greenstone belts of South Africa, Saager et al. (1984, p. 54) recognized "...far reaching similarities in their ore mineralogy with those of the Witwatersrand," and that they "...are primitive forerunners of Witwatersrand gold-uranium placers." Doubtless many of the same geological processes operated in different greenstone belts throughout the Middle to Late Archean, despite the widespread development of supracrustal epicontinental rocks in South Africa much earlier than elsewhere (Pretorius 1981a, 1981b; Eriksson et al. 2006). Stabilization of the crust is also indicated by the occurrence of detrital



FIG. 7.—(A) Rough polished saw section of an ore sample from “B” Reef, Freddie’s Gold Mine, Free State Area (Orange Free State) Goldfield, showing a reworked auriferous concentration of “buckshot” pyrite. Beneath this pyritic layer, there is a black ~2-mm-thick layer of auriferous radioactive carbon (1) resting upon gray quartzite (2). Numerous tiny gold particles (gold colored) occur in the matrix among various types of pyrite, including: layered (3), concentrically zoned (4), rounded compact (5), pyrite overgrown upon rounded compact grain (6), and radial concretion (7). Note large (~3.5 cm diameter) quartz pebble (8). Scale as shown. (Photograph by D.J. Mossman.)



FIG. 7.—Continued (B) More highly polished reverse side of the Freddies Mine sample showing layered pyrite grain (1), large (2.2 cm long) clast in which pyrite, in a manner reminiscent of concretionary growth, cements medium-sized grains of quartz (2), somewhat broken-up basal layer of carbon (3), ~3.5-cm-diameter quartz pebble (4), and basal layer of gray quartzite showing yellow-orange internal reflections (5). Gold-colored reflective mineral is gold. Scale is in centimeters. (Photograph by D.J. Mossman.)



FIG. 8.—(A) Arrows show the contact between the hanging-wall and footwall quartzite in hand specimen of the Basal Reef, Free State Area (Welkom) Goldfield. Contact is marked by millimeter-thick layer (arrows) of carbon flecked with scattered tiny gold particles. Base of sample is blue spray-painted. Scale is in centimeters. (Photograph by D.J. Mossman.)



FIG. 8.—Continued **(B)** Close-up of (cf. Fig. 8A) the contact (arrow) between hanging-wall and footwall quartzite in hand specimen of Basal Reef showing dense waxy-looking quartzite. Waxy appearance is attributable to the micaceous content (mainly pyrophyllite), reflecting perhaps an original clay content. Stope geologists can distinguish the hanging wall from the footwall by subtle difference in texture, an essential aspect of their work to bring development back on the reef in faulted areas. Scale is in centimeters. (Photograph by D.J. Mossman.)

diamonds in conglomerates of the CRG. Thus, from the perspective of the modified placer hypothesis, those processes did not work elsewhere as efficiently to concentrate gold as in the WWR. Then too, Hutchinson and Viljoen (1988) remind us to enquire why such immense enrichment of gold should happen to be uniquely restricted to the WWR. This is a powerful argument against direct detrital derivation of WWR gold from greenstone belts, regardless of any perceived similarities in timing, host lithologies, and mineralogy of deposits.

Pretorius (1982, p. 219) observed, "There are answers for much of what happened within the basin, but only questions for almost all that occurred outside the boundaries." According to proponents of the modified placer hypothesis, the indicated source area of sediments in the basin, however unsatisfactory in terms of potential supply of gold, is usually concluded to be some combination of granite and greenstone (e.g., Hallbauer 1984, Wronkiewicz and Condie 1987, Frimmel et al. 2005b). Like some of the PGM grains, the presence of detrital chromite in the ores is suggestive of such a greenstone source. Saager et al.'s (1984) survey of numerous Archean pyritic conglomerates in southern Africa revealed that constituent chromites all have virtually identical compositions and that they are compatible with chromites derived largely from Archean greenstone complexes.

**Gold:** Morphologically, five types of gold are recognized: (1) very fine-grained particles (average grain size 0.05–0.5 mm), (2) filamentous gold associated with carbon, (3) diagenetically recrystallized gold, (4) gold in secondary quartz veins (extremely rare), and (5) sparse minute inclusions in pyrite grains (Feather and Koen 1975; Hallbauer 1981, 1986). The average overall fineness (800–960) of the gold is usually high (Frimmel et al. 2005a), but figures as low as 650 have been observed (D. Hallbauer, personal communication, 2010). Many other metallic elements are also present.

Different views on gold composition exist. According to Utter (1979), the Ag content of individual gold particles of three morphological types from various reefs in the Klerksdorp area is quite homogeneous, ranging from 7% to 15%. Hallbauer and Utter (1977) and Minter (1990) concluded that fineness can be correlated with sedimentological factors and is thus an indication of source area. Oberthür and Saager (1986) reported that the Hg content of gold from the Carbon Leader Reef ranges from 2.7% to 5.9%, with a mean of 3.1%, but they maintained that the values do not reflect primary conditions (cf. Falconer 2003, Falconer et al. 2006). Reid et al. (1988) found the average composition of gold from the Vaal placer to be Au 90, Ag 8, Hg 2, but noted that their data do not fit a simple model of compositional control according to source or metamorphic homogenization. Thus, it seems that gold composition depends upon the samples analyzed, and upon the place and scale of investigation. Frimmel et al.'s (1993) analysis of "detrital" gold from the Basal Reef placer, Free State Area (Welkom) Goldfield, is held to support postdepositional homogenization on hand specimen scale of 8.9% Ag and 1% Hg. In terms of Au, Ag, and Hg contents, the more definitive results of Hayward et al.'s (2005, p. 39) electron microprobe analyses reveal considerable chemical heterogeneity in (gold) composition on both regional and centimeter scales. Specifically, they documented "a decrease in the amount of heterogeneity in gold composition from the regional scale to smaller scales of mine outcrop, to single thin section and even individual gold grains" (ibid, p. 42) Falconer et al. (2006, p. 525) reported that gold in the Witwatersrand-type Tertiary-aged Belle Brook (New Zealand) deposit is likewise heterogeneous in terms of Au-Ag-Hg composition.

Solution and redeposition of gold (whether or not trapped in a carbon seam) through diagenetic/authigenic or "hydrothermal" processes may account for the perceived heterogeneity of WWR gold. Consider, for example, that layers of carbon located directly upon the paleosurface tend to be much more auriferous than those located higher up in a given reef (Minter 1981, p. P1). Thus, if mobilization

occurred, then it did so after deposition of the carbon such that metallization became more closely confined to the original deposition surface which, according to Minter (1981, p. P2) was "... the lowest and optimum bedload position for gold." Doubtless a second contributing factor to heterogeneity is the presence of resedimented (so-called "granulated") auriferous carbon, which occurs quite commonly in lag gravels.

Gold is found, albeit only in minor quantities, in a number of pre-WWR conglomerates. Reimer (1990) documented a barite-bearing placer carrying a maximum concentration of 1.6 g/t Au together with other heavy minerals from the Fig Tree Group of the Swaziland Supergroup at least by 3.2 Ga. In conglomerates of the overlying Moodies Group, Stupp (1984) found up to 0.5 g/t gold. Auriferous placers have been exploited on a small scale in the Pongola Supergroup (Saager et al. 1984, 1986; Stupp 1984), and Saager and Muff (1986) described a small gold placer deposit from the Uitkyk Formation of the Pietersburg greenstone belt. Its maximum age is 2.901 Ma (De Wit et al. 1993). The Dominion Group conglomerates underlying the WWR were mined essentially for uranium, but they also yielded some gold.

**Uranium:** Uraninites in WWR conglomerates have not been investigated as intensively as the gold. In the main goldfields, uranium, chiefly in round grains of uraninite, usually accompanies gold at every site of deposition, an association present also at other geographic localities in similar quartz-pebble conglomerates (e.g., Elliot Lake, Canada; Jacobina, Brazil). According to Minter (written communication, 1986), uranium occurs in virtually all carbon layers and, like the carbon, is confined within the placer unit. However, one of the main sources of uraninite in the WWR has been the Monarch Reef (Krugersdorp Goldfield), where the carbon-uranium association is not conspicuous.

Descriptions of the uraninites show that the vast majority is made up of extremely well-rounded muffin-shaped grains of 80–150  $\mu\text{m}$  mean diameter. They commonly occur in clusters of up to 20 grains, and, in some cases, as many as six grains are "glued together" like a cellular mass (Koen 1961). Hypidiomorphic grains, which tend to be slightly larger, are also locally present.

Reimer's (1987) compilation of the chemical compositions of the uraninites shows that they exhibit a wide range of compositions, with  $\text{ThO}_2$  ranging from 0.4% to 10.2%. Concerning uraninites not associated with carbon, Feather's (1981) data reveal that there are two distinct populations. The first population (60% of the total) contains 1.5% to 4.5%  $\text{ThO}_2$ , and the second, accounting for about 40%, has values between 5.2% and 10.2%  $\text{ThO}_2$ . In uraninites associated with carbon, the low-Th (0.4%–4.7%  $\text{ThO}_2$ ) fraction accounts for 80% of the grains, whereas the high-Th one (6.2%–8.6%  $\text{ThO}_2$ ) constitutes about 20%. The uraninites of the Main Reef have Th <2.2%, with high U/Th (26.9–40.3). In contrast to this, uraninites of the Vaal Reef have U/Th as low as 8.9.

This bimodal distribution was attributed by Reimer (1987) to recycling of uraninites from the underlying Dominion Group into those of the WWR. Those derived from the Dominion Group are mostly subrounded with mean diameters of 70–130  $\mu\text{m}$ . Their high Th-contents (2.83%–5.8%), the ensuing low U/Th (8.5–16), and their high rare earth element (REE) contents (3.4%) characterize them as varieties usually associated with granitic-syenitic Nb/Ta pegmatites. Columbites and tantalites also occur in the Dominion conglomerates. Reimer (1987) suggested that especially the Vaal Reef and the Basal Reef contain a large contribution of pegmatitic high-Th uraninites recycled from the underlying Dominion conglomerates, as well as low-Th uraninites characteristic of the Main Reef.

According to Pretorius (1991), the answer to the origin of the uraninite lies in the combination of chemical and mechanical recycling. This likelihood is favored by the fact that uraninite (as noted previously) commonly occurs as clusters of round grains, for which

an authigenic origin has been proposed (Liebenberg 1955, Reimer 1975); evidently, solution and remobilization of uranium have occurred, just as they have with gold. At the very top of the sedimentary succession, most of the uranium occurs in brannerite, and lesser amounts of leucoxene—alteration products of uraninite and ilmenite ascribed by Smith and Minter (1980) and Minter (2006) to repeated reworking in an increasingly oxygenated atmosphere. In theory, the oxidation required to produce the freely mobile uranyl ion by groundwater leaching (Finch 1996) could also have occurred by one or more of the following processes:

1. ultraviolet radiation (Benner et al. 2000) ;
2. photolysis of water vapor in the atmosphere, whereby carbon dioxide and water are broken down by light, particularly in the ultraviolet spectrum (Braterman and Cairns-Smith 1987), and photosynthesis, whereby organisms use solar energy to split water, releasing oxygen and reducing CO<sub>2</sub> to organic compounds (these reactions are the basis for generating oxygen from CO<sub>2</sub>, and led up to the Great Oxygen Event, which followed WWR sedimentation by ~200 million years); and
3. microbial activity (Wadden and Gallant 1985).

A large proportion of uranium in the ore may have been precipitated by bacterial action. Several common species (dead or alive) are renowned for their ability to accumulate uranium intracellularly and/or extracellularly (Strandberg et al. 1981, Mann and Fyfe 1985). Also, apart from microbes, organic materials with functional groups (e.g., humic acids) bind with uranium and, with pH change, lead to its precipitation. As Leventhal (1985, p. 11) noted, either hot or cold water can transport a wide range of uranium complexes, and either organic matter alone, or sulfides alone, can lead to the reduction of organic-complexed uranium. Bearing in mind Fyfe's (1985, p. 5) observation that "...gold behaves rather like uranium, being dissolved under acid oxidizing conditions and deposited under more reducing or alkaline condition," one might reasonably conclude that rather similar environmental conditions as well as the mixed assemblage of basic to granitic source rocks may have contributed to the association of gold and uranium. Certainly, judging from the presence of detrital pyrite in the Belle Brook deposit, New Zealand, those conditions need not have been anoxic. The phenomenon is perhaps best ascribed to redox reactions involved in photosynthesis and photolysis.

Giusti et al. (1986) reported Pb isotopic compositions of allogenic gold, pyrite, and sphalerite from the Basal Reef, indicating an age of ca. 3.2 Ga. However, as outlined by Köppel and Saager (1974) and Frimmel (2005, p. 14), and disregarding problems inherent in radiometric dating these constituents using U-Th-Pb isotopic systems, it is appropriate to recall that there are after all, at least five types of WWR pyrite (Hallbauer 1986), three major morphological types of gold (Utter 1979), and several types of PGMs, some of which are texturally and mineralogically complex (Reimer 1979). Bearing this in mind, it is scarcely likely, for example, that "buckshot" pyrite or carbon-hosted filamentous gold will register pre-WWR dates, despite reports (Kirk et al. 2001, 2002) that some gold and pyrite predate WWR sedimentation.

**Pyrite:** Pyrite is present in WWR reef at an average of about 3% to 5% (Ramdohr 1958; Saager 1970, 1981). The main varieties recognized by Hallbauer (1986) (with their proportions of total pyrite) are: (1) allogenic (detrital) (30%–70%), (2) syndimentary (with highest gold content of any pyrite) (10%–15%), (3) pseudomorphs and replacements of various pebbles (<10%), (4) authigenic (5%–40%), and (5) pyrite as small secondary veins (<10%). Texturally, syndimentary pyrite encompasses several categories, including: oolitic, radial concretionary, mudball, and framboidal. Simplifying,

Hutchinson and Viljoen (1988) recognized three main *textural* types, namely, allogenic (rounded compact), allogenic (rounded porous), and authigenic (idio- to hypidiomorphic), all of which are auriferous. However, no matter what the basis for classification, some types, such as porous pyrite, although usually occurring as "buckshot," are occasionally found up to 60 mm in diameter. This latter type could not possibly have undergone appreciable transport. Saager (1981) demonstrated major geochemical differences between the pyrites formed in situ and "primary" and detrital WWR pyrite samples, commenting on the remarkably homogeneous trace-element values (and the high Ni, Pb, and Co contents) in the porous pyrites.

For the record, analysis (by instrumental neutron activation analysis, atomic absorption analysis, and electron microprobe) of trace elements in the authigenic and allogenic pyrite of various WRG and CRG conglomerates reveals gold contents over 9 ppm in "economically interesting conglomerate horizons" together with hundreds of parts per million Co, Cu, Pb, and As (Stupp 1984, p. 118). Hallbauer et al. (1978) reported values up to 1400 ppm in "mudball" pyrite. Hutchinson and Viljoen (1988, p. 160) gave the average gold content of authigenic pyrite of 21 "various mineralized reefs" as 166.8 ppm (units should be ppb—R. Viljoen, written communication, 2010). Clearly, there is an intimate genetic association of pyrite with metallization. Although there has been little detailed accounting/quantification of those elements other than Au and U in the mix, the data testify to a manifestly impressive geochemical imprint along the edge of the WWR basin. Calculation of abundance factors of minerals and metals relative to gold in WWR ores (and of trace elements in pyrite) thus remains a challenging assignment.

**Platinum group minerals (PGM):** Platinum group minerals in the conglomerates are undoubtedly mostly of basic to ultrabasic provenance, as is also true of chromite. The platinum group elements (PGEs) closely follow gold through metallurgical processes of recovery and refining, fingerprinting bullion by trace amounts of Ru, Rh, Pd, Os, Ir, and Pt. The presence of these elements distinguishes WWR gold from that of greenstone belts in Zimbabwe and Mozambique (Goldstuck 1996). Relative proportions of the various PGEs do not change significantly throughout the basin. Reimer's (1979, p. 296) compilation shows them present at 0.6 to 9.4 ppb in the Main Reef and Bird Reefs throughout the basin. In the Kimberley Reefs of the East and Far East Rand Goldfields, values as high as 2.2 ppm have been observed. Recovery efficiency is estimated at 50% to 60%. Most PGM grains are alloys of the Os-Ir-Pt-Rh series, and the Pt-Fe series and PtAs<sub>2</sub> (sperrylite) are found in lesser but about equal amounts. PGMs throughout the basin tend to lack Pd, possibly because of some predepositional process, according to Reimer (1979). Most are described as detritals, but some occur as inclusions in, and overgrowths on, alloys and other minerals. Of the dozens of PGMs recorded by Reimer (1979), over 10% are concentrically banded alloys, among them, complex nondetrital intergrowths of PGE and Au present in what Ramdohr (1958) referred to as a coal-like substance. Cousins (1973) showed that some platinoïd grains, of mainly osmiridium composition, are mature alluvials associated with gold and other heavy minerals.

Employing osmium isotopic systematics to heavy mineral concentrates from various WWR mines, Hart and Kinloch (1989) observed that some Os-Ir grains have model ages older than 3.02 Ga. This finding supports the observations of Cousins (1973) and tallies with the results of Kirk et al. (2001, 2002) concerning the putative age of some "detrital" gold grains in the WWR (although this can scarcely be expected to apply to, say, the filamentous gold contained in carbon). Not surprisingly, an appreciable spread in ages was recorded among osmiridium grains by Hart and Kinloch (1989), who concluded that not all of the osmium was contributed by detrital grains. While some grains have model ages older than the maximum age of the WWR (3.02 Ga, according to Armstrong et al. 1991), the youngest grains, at 2.75 Ga,

strain the detrital hypothesis. We supply data for one unequivocally detrital grain from Ramdohr's collection (Schidlowski 1970, plate 22–2). This grain has an  $^{187}\text{Os}/^{186}\text{Os}$  ratio of  $0.8983 \pm 4\%$  (1s) and yields a model age of ca. 2.86 Ga, according to the plot provided by Hart and Kinloch (1989, their Fig. 1, p. 1654). In this example, the osmium evidently will have had a residence time in a crustal reservoir prior to isolation in the WWR. However, osmium mobility is evidenced by the common rimming of osmiridium on various Os-Ir-S-As species (Cousins and Kinloch 1976, Feather 1976). Melcher et al. (2005) documented several processes in geologically recent alluvial sediments that have contributed to chemical and textural modifications of detrital PGMs.

### *Models and Microbes*

As one of Earth's oldest large basins, the WWR succession preserves ore at several stratigraphic levels within a great thickness (>7000 m) of quartz arenite, shale, and BIF. In detail, it hosts a complex suite of economically important minerals, including gold, uraninite, sulfides, and PGMs, in three pyrite-rich sedimentary facies. Mineral zoning, normally a characteristic feature of basins in which sulfide minerals form and where precipitation is governed by chemical facies, is conspicuously absent. Deposits in the WRG and the CRG do not occur haphazardly in the basin but lie along preferred stratigraphic horizons in quartz arenites. Most important, reefs are situated above major transgressive erosional breaks. These horizons are "more mature" in that they are notably even more siliceous than their highly siliceous metasedimentary host rocks. The high tonnages of Au and U produced stem in large measure from the spectacular concentration of these elements in thin extensive carbon reefs at the Main Reef and Bird Reef horizons. The tremendous buildup of gold over narrow widths, in places only a few millimeters thick yet extending over tens of square kilometers, is an indication of extremely efficient processes of metal concentration, unrivaled in any other goldfield in the world. The model is simple. Sea level rises, sedimentation slows, and syngenetic gold is precipitated out in microbial mats in shallow water or in situ, along with pyrite in "stagnant" gravel deposits onshore (see Fig. 5A–C). Then, sea level falls, reactivating the entire sedimentary system, reworking the gold into more "classic" placer-type deposits. The extent of reworking becomes dispersive through time, of course, as this relatively fine-grained gold is not readily concentrated in most erosional environments.

The concentration of gold within conglomerate beds by the bedding-parallel flow of hydrothermal fluids over tens of kilometers envisaged by revisionists (e.g., Phillips and Law 2000) is an unsatisfactory model, as outlined by Mossman et al. (2008) and by Frimmel and Minter (2002, p. 34); nor is Hutchinson and Viljoen's (1988) auriferous pyritic exhalite model—held as the source of ironstones and placer gold in the WWR succession—particularly apt. Analogous to modern seafloor hydrothermal systems, that hypothesis envisages hydrothermal fluids discharged around the edges of a fault-bounded shallow-marine or continental basin. Although it might accommodate several puzzling aspects of WWR geology, the presence of Lake Superior-type iron formation, at least in the lower half of the stratigraphic column, is not compatible with the exhalite model. The low gold content in the ironstone and shale is more likely due to element dispersion than to an exhalative process. Also, chemosynthetic and photosynthesizing organisms will have contributed requisite oxygen for BIF formation in the WWR succession.

However, no matter the particular model, or combination of models invoked, a supply of aqueous metal-rich fluid of "hydrothermal" chemical composition seems required to have supplied such a large amount of metals. As Handley (2004, p. 14) states: "The real crux of the matter is to arrive at a means of concentrating this gold and presenting it in a series of deposits within a single geological

succession." It is well established that hot springs are quite capable of incubating gold deposits at grades (and quantities) equal to that of the WWR. With respect to water-dominated geothermal systems such as those on North Island, New Zealand, Brown (1986, p. 982) concluded that the gold contents of the deep fluid of the Ohaaki geothermal field "...obviate the need to invoke special chemical considerations or specially pre-enriched (sic) host rocks in order for a geothermal system to generate a precious metal ore deposit." Consider, for example, the *average* of 1.5 ppb Au (a figure well below the 10.5 ppb solubility predicted from thermodynamic values), which occurs in the deep fluid of hydrothermal production wells at Ohaaki (Brown 1986). Let us instead choose a lower number, say 0.3 ppb, thereby allowing for inefficiency in precipitation. Thus, at one-fifth of the above *average*, and given a total flow rate for the Ohaaki field of 38,400 t/day (Brown 1986, p. 982), ~0.01152 kg/day of gold is precipitated. In 1 million years, a total of ~4200 mt Au will have accumulated, an estimate which approaches the amount of gold recovered from the Main Reef, for which Sanders et al. (1994) list the figure as 4800 t.

Although there are no recognized remnants of possible feeder pipes to geothermal systems either within or bordering the WWR basin, it is worth looking closer at some of the geochemistry involved. The Ohaaki fluids transport Au as the hydrosulfide complex  $\text{Au}(\text{HS})_2^-$  under conditions that doubtless (at least normally) lead to deposition of much gold due to adiabatic boiling. However, in these geothermal systems, even at near-surface temperatures of 100° C and less, bisulfide complexing is dominant, keeping the gold in solution. According to Lewis (1982), even at 100° C and less, the waters will have remained highly undersaturated in gold, and these conditions would permit relatively long-distance transport of dissolved gold. Furthermore, the changes in temperature and pH in this situation are far less important mechanisms than oxidation in governing the precipitation of gold. These conditions are precisely those favored by facultative aerobic prokaryotic microorganisms, bringing to mind the scenario described by Reimer (1984), Mossman and Dyer (1985), Southam and Beveridge (1994), and Mossman et al. (1999), whereby transport of gold occurred as a solution or colloid, stabilized by humic acids or by sulfur cycle intermediates, with biologically induced precipitation of gold ensuing as a result of oxygen produced in localized environments by microbial communities. In this setting, the strongly electronegative nature of gold would also result in its attraction to associated clay particles. Brown (1986, p. 982) suggested a role for the transport of  $\text{Au}^0$  as a stable colloid in the Ohaaki field. This could come about through electron transfer and weak bonding between gold atoms, leading to negatively charged clusters forming colloids (Williams-Jones et al. 2009). Indeed, colloidal suspensions have now been confirmed as a powerful mechanism for gold transport in nature (Hough et al. 2008), as is the synthesis of nanoparticles of gold by filamentous cyanobacteria from thiosulfate and chloride complexes (Lengke et al. 2006a, 2006b; Lengke and Southam 2005, 2007). The results of biochemical processes, supplementing low-temperature chemistry of aqueous brines, could thus account for concentration of a substantial portion of the syngenetic gold in Earth's greatest repository of gold.

In recent years, the potential contribution of microbial communities to concentrating Au and U in ore deposits has come into sharp focus. The Late Archean and Early Proterozoic represent, after all, the age of prokaryotes. An especially relevant factor in the present context is the role that microbial communities can and do play in geothermal settings (Beaumont and Foster 1988, Dobson et al. 1988). Their functions are diverse (Reith et al. 2007). Metabolic pathways of different species allow for a wide range of electron acceptors for respiration, in addition to oxygen. Microbes can serve as catalysts, speeding up otherwise slow redox reactions in order to achieve a particular geochemical impact. In so doing, a microbial community may also create specific microbial fingerprint(s), essentially molecular fossil evidence reflecting the nature of the geochemical processes involved (e.g., Waldbauer et al.

2009). The solubility of gold by methylation and its ability to readily form complexes with  $\text{Cl}^-$ ,  $\text{HS}^-$ ,  $\text{S}_2\text{O}_3^{2-}$ ,  $\text{CN}^-$ , and  $\text{SCN}^-$ , etc. (Lengke and Southam 2005, 2007), and the interaction of bacteria with metallic ions in solution (Strandberg et al. 1981, Beveridge and Fyfe 1985, Fyfe 1985, Kuyucak and Volesky 1989) are also major factors in favor of the feasibility of the geochemical precipitation of much of the WWR gold. Not surprisingly,  $\text{Au}^0$  forms the strongest complexes with  $\text{CN}^-$ , a degradation product of organic matter. Results of in vitro experiments (Grosovsky 1983; Dexter-Dyer et al. 1984; Mossman and Dyer 1985; Dyer et al. 1988, 1994; Southam and Beveridge 1994) leave no doubt about the ability of prokaryotes to accumulate gold to even percentage amounts and to contribute to the genesis of gold deposits. As noted earlier, much the same argument also applies to uranium (e.g., Strandberg et al. 1981).

## DISCUSSION AND CONCLUSIONS

Smith and Minter (1980, p. 12–13) concluded that fluvial processes in the WWR braided stream environment were not competent to generate large concentrations of gold, but that some unknown processes dumped the putative “detritals” into the margins of the basin of deposition. Recall too, Hallbauer and Utters’s (1977) documentation of short-range (<40 km) transport of detrital gold grains extracted from the conglomerates by digestion in hydrofluoric acid. Concerning source rocks, Minter (1990, p. 199) summed up the overall dilemma: “The main problem lies in accounting for the sheer quantity of not only the gold but also of the pyrite and uraninite in the placers.” Indeed, any case against the wholesale detrital derivation of gold in particulate form from the hinterland that is formulated on the basis of the immense tonnage involved grows with the realization that even supposing that ~39,000 t of gold remain to be recovered (Frimmel and Minter 2002, p. 17), there likely remains at least an equivalent amount of unpayable gold “reserves” in excess of the estimated  $2 \times 50,000$  t ultimately recoverable. Other workers (e.g., Reimer 1975, 1984; Pretorius 1991) have also argued that there is too much particulate gold in the WWR conglomerates to have been entirely derived by detrital processes from a “fertile” hinterland rich in hydrothermal gold deposits. In contrast, Loen (1992) calculated that the total gold embedded in the WWR could easily have been derived by weathering of a source exhibiting no unusual concentrations of gold. Note, however, that such gold would be present mainly as chemically dissolved gold and/or as minute inclusions in various minerals. This “chemical” gold—transported in solution or as colloids to the edge of the WWR basin—would have to have been transformed into grains of a size amenable to mechanical emplacement as a placer. The most likely loci of transformation would have been shallow-water environments along the basin edge in which bacteria thrived. These would be gold/carbon associations that at times extended far into the basin itself. Where subsequently reworked, whether within or outside of the preserved boundaries of the basin, these zones will have contributed significantly to metallization in the conglomerate placers by normal sedimentary processes. Recognition of this feature substantially eases the problem of accounting for the total mass of WWR gold.

Results of detailed studies (e.g., Viljoen 1968, Pretorius 1981a) have shown that the sediments and their metal content are related to drainage patterns that have persisted through >7000 m of stratigraphic section and numerous regional unconformities. This remarkable feature suggests that metalliferous aqueous fluids (whatever their origin and levels of concentration and temperature) reached depositional sites via streams flowing into the basin—in effect a process of chemical recycling of available metals through precipitation, and subject in part to mechanical recycling by various fluvial agencies. It remains only to superimpose a regime of intense chemical weathering to kick start the process, with follow-up late-stage microbially mediated metallization. Note that the key horizons are carbon-bearing reefs within the ~500-

m- to 1000-m-thick Johannesburg Subgroup, where the Main Reef in the Rand and West Wits Line (Carletonville) Goldfields and the Bird Reef in the Klerksdorp Area (Klerksdorp) and Free State Area (Welkom) Goldfields have together accounted for over 84% (two thirds of it from the Main Reef) of all WWR gold that has been produced (Sanders et al. 1994).

Diverse scenarios have been envisaged as providing hydrothermal activity in the equation of WWR metallization. Few workers will dispute that decimeter-scale “hydrothermal” mobilization of gold has been superimposed upon local and regional metamorphic effects (Ramdohr 1958, Frimmel et al. 1999, Volbrecht et al. 2002, Frimmel et al. 2005b, Hayward et al. 2005). However, the perceived results of that mobilization are for the most part indistinguishable from those achieved by diagenetic/authigenic processes (Volbrecht et al. 2002). The “revisionist” school (e.g., Phillips and Myers 1989, Barnicoat et al. 1997) would account for the gold by penetrative bedding-parallel process of hydrothermal flow. However, no discernible migration of metals has been shown to occur beyond the sedimentary limits of deposition, except for minor quartz veins and along dike contacts. Hayward et al. (2005) maintain that widespread circulation of auriferous fluids did not occur except for minor fluid-poor mobilization of gold under metamorphic conditions as a result of the Vredefort meteorite impact. Thus, neither the hydrothermal hypothesis nor the modified paleoplacer hypothesis (see also the summary of arguments in Frimmel et al. 2005b, their Table 1, p. 13) can satisfactorily account for the metallization. One is reminded at this point of Sherlock Holmes’ old axiom: “. . .when you have excluded the impossible, whatever remains, however improbable, must be the truth” (*The Adventure of the Beryl Coronet*). What remains here as the only viable explanation is that a substantial proportion of gold and uranium in WWR ores resulted from chemical precipitation—as concluded by De Launay (1896) at a time prior to recognition of Witwatersrand “carbon.”

The role of microbial activity and the consequent chemical and biochemical concentration of gold will have begun very early. The carbon-gold association appears already in conglomerates of the Moodies Group of the Swaziland Supergroup (3.25 Ga), where Stupp (1984) recorded the occurrence of flyspeck carbon, remarking on the absence of columnar-type carbon in pre-WWR conglomerates; gold concentrations there are about 0.5 g/t. Mossman et al. (2008) mentioned one auriferous carbon seam preserved in a quartz-pebble conglomerate in the 3.01 Ga Dominion Group. In the Pongola Supergroup (2.9–3.2 Ga), Stupp (1984) also observed the flyspeck association, and in a conglomerate of the Nsuze Group (the lower part of the Pongola Supergroup), he observed a 3-mm-thick carbon seam characterized by a high concentration of gold particles (usually >15  $\mu\text{m}$ ) up to 110  $\mu\text{m}$  in diameter. Then, as Walsh and Lowe (1985) predicted, and as Noffke et al. (2003) and Noffke (2008) have since confirmed, remnants of algal mats are well preserved in these ancient (Pongolan) metasedimentary rocks. Concentrations of gold at such horizons can in part be explained by baffling (see also Utter 1979, p. 33), although microbial communities will have promoted biochemical precipitation and adsorption of gold along particular horizons during sedimentation. A remarkable contemporary example of the baffling action of algae was brought to our attention by K. Hein (written communication, 2008), who describes tiny grains of gold entrapped in modern algae near the Sharks Gully mine in the Coongan greenstone belt of the Pilbara Craton in the Fortesque Supergroup, an Australian equivalent of the Witwatersrand Supergroup.

The picture of a modern geothermal system was summoned earlier to illustrate the extraordinary ore-forming potential of metals in solution at relatively low temperature at Earth’s surface. The thing is, however, that volcanic degassing could equally well have served the purpose as a major driver of WWR metallization. For WWR metallization, all four aspects critical to ore genesis (White 1968) are

satisfied, namely: aggressive chemical weathering (*source*) yielded metal-enriched aqueous solutions (*concentration*), which accessed (*migration*) restricted environments of shallow-water placers. The result was chemical and biochemical *precipitation* of a complex assemblage of minerals and metals superimposed upon any and all residual refractory minerals, where the whole package was ultimately subject to various surficial and diagenetic geological processes prior to lithification. In view of the performance record of modern analogues, organically supported processes played important roles in many of those processes.

In summary, syngeneses resulted in the superimposition by chemical precipitation of gold, uranium, and pyrite and associated elements Co, Ni, Cu, Zn, Pb, and As along the WWR basin edge in thin, areally extensive deposits upon chronostratigraphic unconformities in notably unmineralized siliciclastic sediments. The alluvial character of a large part of the ore resulted from reworking in fluvial environments. Syngeneses goes further than the paleoplacer hypothesis (however modified) or the hydrothermal hypothesis, *sensu stricto*, in accounting for the following:

- The braided stream environment of most WWR deposits was not competent to generate the observed large metal concentrations.
- Some conglomerates were mineralized, whereas others of similar composition in the vicinity were not.
- Reefs that formed as separate lobes of conglomeratic sediment, some oligomictic and others polymictic, nevertheless have similar complex suites of ore minerals.
- The controlling feature of the ore begins and ends with its relation to erosion surfaces.
- Those erosion surfaces are chronostratigraphic unconformities left virtually unchanged over long time intervals, which served as substrates for the growth of microbial mat communities.
- Over 40% of WWR gold is spatially associated with carbon reef seams, the remnants of microbial mat communities.
- The microbial mats leading to the carbon reefs formed and flourished on paleosurfaces (unconformities, disconformities, shallow erosion channels) during pauses in sedimentation succeeded directly in time by accumulation of clean quartz arenite.
- The carbon reefs and other principal reefs are only a few centimeters thick, yet they extend for tens of kilometers.

Given the central role of intense chemical weathering in the early emergent Kaapvaal Craton, the key confluence of favorable ore-forming conditions was completed with the blooming of microbial mats during prolonged hiatuses in sedimentation. Geochemical concentration of gold was aided and abetted biochemically through intra- and/or extracellular concentration by (living) microbial activity, and passive absorption by activated carbon derived from dead microbes. Passive baffling of wind/water-transported micronuggets and flakes of gold will have added to the efficacy of the overall process. Thus, although a general upward increase in the concentration of gold doubtless resulted in part from erosional recycling, a large proportion of the gold was emplaced under relatively low-temperature, chemically aggressive atmospheric conditions. This occurred notably at several carbonaceous horizons along the preserved margin of the WWR basin, depending upon the availability of aqueous metalliferous fluids at chronostratigraphic unconformities coincident with stillstand of the land surface degradation and consequent proliferation of microbial growth. Likewise, some of the gold precipitated by this process outside of the preserved basin would have been reworked and emplaced as detritus within the conglomerates with or without originally associated carbon.

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