Available online at www.sciencedirect.com
ScienceDirect
EPSL
Earth and Planetary Science Letters xx (2007) xxx–xxx
www.elsevier.com/locate/epsl

Earth's continental crustal gold endowment

H.E. Frimmel *

Division for Geodynamics and Geomaterials Research, Institute of Geography, University of Würzburg, Am Hubland, D-97074 Würzburg, Germany

Received 18 May 2007; received in revised form 28 September 2007; accepted 17 November 2007

Editor: C.P. Jaupart

Abstract

The analysis of the temporal distribution of gold deposits, combined with gold production data as well as reserve and resource estimates for different genetic types of gold deposit, revealed that the bulk of the gold known to be concentrated in ore bodies was added to the continental crust during a giant Mesoarchaean gold event at a time (3 Ga) when the mantle temperature reached a maximum and the dominant style of tectonic movement changed from vertical, plume-related to subhorizontal plate tectonic. A magmatic derivation of the first generation of crustal gold from a relatively hot mantle that was characterized by a high degree of partial melting is inferred from the gold chemistry, specifically high Os contents. While a large proportion of that gold is still present in only marginally modified palaeoplacer deposits of the Mesoarchaean Witwatersrand Basin in South Africa, accounting for about 40% of all known gold, the remainder has been recycled repeatedly on a lithospheric scale, predominantly by plate-tectonically induced magmatic and hydrothermal fluid circulation, to produce the current variety of gold deposit types. Post-Archaean juvenile gold addition to the continental crust has been limited, but a mantle contribution to some of the largest orogenic or intrusion-related gold deposits is indicated, notably for the Late Palaeozoic Tien Shan gold province. Magmatic fluids in active plate margins seem to be the most effective transport medium for gold mobilization, giving rise to a large proportion of volcanic-arc related gold deposits. Due to their generally shallow crustal level of formation, they have a low preservation potential. In contrast, those gold deposits that form at greater depth are more widespread also in older rocks. This explains the high proportion of orogenic (including intrusion-related) gold (32%) amongst all known gold deposits.

The overall proportion of gold concentrated in known ore bodies is only 7×10^{-7} of the estimated total amount of gold available in the continental crust. This is less than the solubility of Au in common crustal fluids. A high potential for the existence of voluminous, hitherto undiscovered, gold resources may thus be inferred.

Keywords: gold; lithosphere; Archaean; Witwatersrand; recycling

1. Introduction

Gold, for its high resistance to chemical reaction, high conductivity and ductility, and its rarity has been one of the most sought after commodities for at least 7000 years. Total historic gold production is estimated (based on Gosselin and Dube, 2005, updated) at approximately 183,000 metric tons (t). This gold has been mined from a variety of deposit types that range in age from Archaean to Recent. In spite of its generally very low reactivity, gold is locally concentrated in ore bodies that reflect enrichment by a factor of 10^4 relative to background levels. The large variability in the geological settings of gold deposits implies transport and concentration of gold by magmatic, hydrothermal, and sedimentary processes. The factors that control the concentration of gold at certain sites in the Earth's crust to deposit level as well as the ultimate source of the gold in the various deposits remain, however, topics of debate (e.g. Bierlein et al., 2006; Groves et al., 2005).

Even on a scale beyond that of ore deposits, gold is distributed unevenly in Earth. The modern continental crust is, with an average Au concentration of 1.5 μg kg^{-1}, enriched by a factor of 1.57

* Tel.: +49 931 8885420; fax +49 931 8884620.
E-mail address: hartwig.frimmel@uni-wuerzburg.de.

0012-821X/$ - see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.epsl.2007.11.022

relative to primitive mantle (Palme and O'Neill, 2005; Rudnick and Gao, 2005). Crustal gold represents, however, only a miniscule proportion of the total amount of gold in Earth. As chondritic meteorites contain two orders of magnitude more gold than silicate Earth (McDonough and Sun, 1995), it has been concluded that the vast majority of gold partitioned into the core during early differentiation of the planet (Wood et al., 2006). Following that argument, 98% of Earth's gold should be present in the core. As effectively all known gold deposits are located within the more accessible continental crust, it is this portion of the planet that shall be of interest here.

By analyzing data on global gold production from more than 1800 deposits/occurrences, the relative significance of different types of gold deposit and the distribution of gold deposits over geologic time, the principal mechanisms of gold distribution in Earth's continental crust will be discussed and the amount of gold available therein evaluated. Previous analyses of this kind focused on orogenic gold (Goldfarb et al., 2001, 2005), the seemingly most important type of gold deposits in terms of global distribution. However, they did not take into full account the by far largest known gold depository in the Witwatersrand Basin of South Africa because of conflicting hypotheses concerning its genesis and age. Here it will be shown that the vast majority of known gold in the continental crust was added to the crust already by approximately 3.0 Ga. This finding has some major implications not only on our understanding of the first-order controlling factors on the spatial and temporal distribution of gold deposits but also on models for Archaean crustal growth in general.

2. Relative significance of different gold deposit types

No uniform classification scheme exists for gold deposits, which certainly reflects some remaining uncertainties in our understanding of these deposits but also a bias by different researchers and interest groups. A given deposit may be classified according to the host rock (e.g. sediment-hosted), or according to a preferred genetic model (e.g. orogenic deposit), the classification may emphasize a specific paragenetic association (e.g. iron oxide copper-gold deposit) or it may be based on the comparison with a large prototype (e.g. Carlin-type). The fundamental weakness of the existing classification schemes is that different deposit types do not necessarily exclude each other. For example, various genetic models have been proposed to explain one of the most important styles of gold mineralization, i.e., Carlin-type deposits (see review by Cline et al., 2005): these include comparisons with both orogenic deposits, involving metamorphic and magmatic fluids, and intrusion-related deposits. Similar ambiguities exist in the case of some of the world's largest know gold deposits, such as the giant deposits in the Tien Shan gold province (Muruntau, Vasilkovskoye, Amantaitau, Kumtor), Sukhoi Log or Telfer. While some workers prefer an orogenic model for all of these deposits (Goldfarb et al., 2005), there is increasing evidence for at least some of these deposits to be intrusion-related (e.g., Mao et al., 2004; Morelli et al., 2007). Similarly, many other, smaller deposits that have been classified as orogenic are increasingly linked to intrusions, thus blurring the distinction between orogenic and intrusion-related deposit types. A special case is the Witwatersrand Basin in South Africa — by far the single most important gold province worldwide. Genetic models proposed for the Witwatersrand gold range from intrusion-related to orogenic and palaeoplacer deposits (for a discussion of contrasting views see Muntean et al., 2005). Today compelling evidence for a palaeoplacer model exists (see review by Frimmel et al., 2005), which has profound implications on our understanding of the global gold distribution and the principal mechanisms that led to the concentration of this noble metal at certain sites in the Earth's continental crust.

As basis for this study serves the analysis of historic gold production data and current reserve and resource estimates (Gosselin and Dube, 2005; Handley, 2004; Porter and Amey, 2003; Raw Materials Data© 2007, unpubl. own data). As with most commodities, the last few decades have seen higher production rates than ever before. Almost one third of the global historic gold production stems from the last two decades alone. Production statistics over the past decades are more readily available and the period 1984–2006 is therefore taken as reference for the assessment of the relative significance of different gold deposit types for the overall gold production.

Here we focus on the principal Au sources and crustal-scale Au-enrichment processes. Instead of distinguishing between dozens of different deposit types that emphasize district-scale differences, only a few types of deposit, largely grouped according to genetic relationships, are differentiated and their plate tectonic position(s) summarized in Fig. 1. For examples of the most prominent representatives of each type see Table 1.

From the comparison of past production and known reserves and resources from each type (Fig. 2) it becomes apparent that the placer and orogenic (including intrusion-related) gold deposits are by far the two most important types. While the orogenic and intrusion-related deposits include some of the largest known individual deposits, such as Muruntau (Uzbekistan), and are spread widely both spatially and temporally (Goldfarb et al., 2001), almost all of the known placer deposits (91%) are located in a single gold province, the Witwatersrand Basin. This Mesoarchaean sedimentary basin accounts for about 40% of all known gold and thus represents the by far largest known gold depository. Most of the pre-modern gold production, notably from the Arabian–Nubian Shield, the European Variscan belts and the Yilgarn Craton, was probably derived from orogenic gold deposits, but no precise production data are available. For this study, the minimum estimates for past production until 2000 are taken from Goldfarb et al. (2001) and complemented with available data for the past six years. Note that the genesis of some examples that are assigned here to the orogenic type is debatable (e.g., Sukhoi Log).

Deposits hosted by both oceanic and continental volcanic arcs are the next most important hosts of gold (Fig. 2). These include porphyry Cu deposits, often associated with Mo when located in a continental arc, some intrusion-related and skarn deposits and, more distal, sediment-hosted deposits. Although highly variable in the style of mineralization, this type of gold is sourced in magma and related magmatic fluids that are typically derived from the partial melting of lithospheric mantle above

Please cite this article as: Frimmel, H.E., Earth's continental crustal gold endowment, Earth Planet. Sci. Lett. (2007), doi:10.1016/j.epsl.2007.11.022

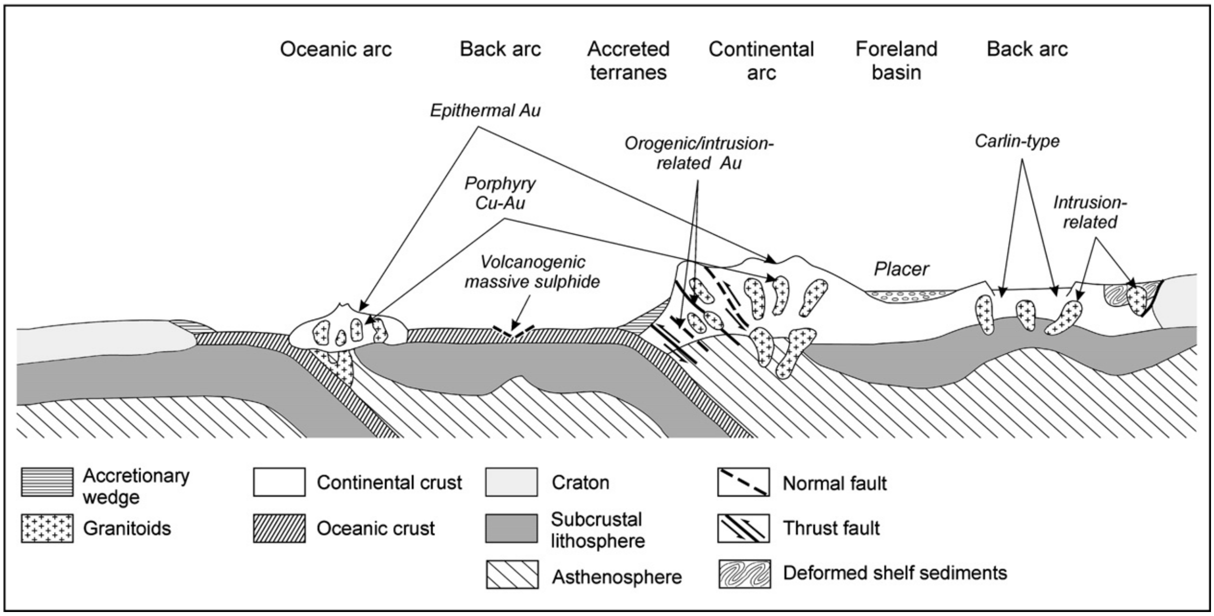

Fig. 1. Sketch illustrating the most common lithosphere-scale environments for the formation of the principal gold deposit types as discussed in the text (modified from Groves et al., 2005).

subduction zones. Though low-grade, porphyry Cu–Au systems can attain large tonnage. The equivalent expression of such mineralizing systems at very shallow levels or on the surface of volcanic arcs is in the form of epithermal deposits. With a total of 17% volcanic arcs contain an important proportion of all known gold, mainly as epithermal and porphyry Cu–Au deposits.

In contrast to the orogenic and volcanic arc-related deposit types, Carlin-type deposits (for a review see Cline et al., 2005) formed in an extensional rather than compressional stress field behind a continental arc. They are typically replacement bodies hosted by sedimentary rocks at the interface with less permeable strata. Although the temperature of ore formation is lower than in orogenic deposits, the fluid chemistry is similar to that of deep metamorphic fluids as in orogenic deposits. In contrast to orogenic deposits, Au is present as solid solution or submicron-size particles in disseminated pyrite, marcasite or arsenopyrite. While the age of these deposits, all of which are in the type area of Nevada, is now well constrained within a very narrow time window in the Eocene, after orogenic activity had provided steeply dipping fluid pathways, the genesis remains enigmatic (Cline et al., 2005). Gold mineralization has been ascribed to meteoric water circulation (Emsbo et al., 2003), emplacement of shallow plutons (Henry and Ressel, 2000), and the ascent of deep metamorphic and magmatic fluids (Hofstra and Cline, 2000).

The remaining types of gold deposits all represent only a very small proportion (≤1%) of the total known gold. These include ironoxide–copper–gold (IOCG) deposits, which occur in a variety of settings, including volcanic diatremes, and which are still poorly understood (Williams et al., 2005). Volcanogenic massive sulphide (VMS) deposits that precipitated on or below the sea floor by hydrothermal discharge related to submarine, typically bimodal volcanism in rifted-arc and incipient back-arc basins or back-arc rifts can contain economic Au contents (Hannington et al., 1999) but in total they contribute not more than 1% to the total known gold amount. Even less Au is hosted by mafic/ultramafic igneous complexes in which ore-grade concentrations of platinum group elements or Ni and Cu is accompanied by Au-enrichment (e.g. Bushveld Complex in South Africa, Great Dyke in Zimbabwe, Norilsk in Russia). Similarly, syn-sedimentary gold deposits play only a very subordinate role. Gold-enrichment in sedimentary-exhalative (SEDEX) deposits is to be distinguished from elevated Au contents in certain marine sediments. Archaean banded iron formations typically have elevated Au contents (e.g. Meyer and Saager, 1985), though some of the gold deposits in this environment (e.g., Bounty, Australia, and Kalahari Goldridge, South Africa) are believed to be epigenetic. Highly carboniferous shales, such as Early Cambrian black shales on the Yangtze Platform (China), the Permian Kupferschiefer (Poland, Germany), and possibly also the Neoproterozoic shales at Sukhoi Log (Russia) contain elevated Au contents. Although some workers argued for an epigenetic origin of the metal-enrichment in these shales, a case for a seawater origin of these metals, including Au, has been made (Lehmann et al., 2007). Recent experiments (Emsbo and Koenig, 2007; Williams-Jones and Migdisov, 2007) have shown the potential for Au transport by petroleum. Although the overall significance of bituminous deposits as hosts of gold may seem very limited at this stage, the enormous volume of such deposits could represent a formidable, so far unrecognized Au resource in the future.

3. Constraints on the continental crustal gold budget

Of main concern here, leaving aside the core as principal gold reservoir, is the question as to how and when gold was concentrated into the continental crust and ultimately into ore deposits. Typical Au concentrations of shales are similar to those in various magmatic rocks (compare Connors et al., 1993;

Please cite this article as: Frimmel, H.E., Earth's continental crustal gold endowment, Earth Planet. Sci. Lett. (2007), doi:10.1016/j.epsl.2007.11.022

Table 1
Known amounts of gold from principal classes of gold deposits, with major examples (>1000 t Au)

Deposit/district/province	Country	Plate Tectonic setting	Age	Au (t)	Grade (g/t)
Placer deposits (including modified palaeoplacer), total				104 503	
Witwatersrand Basin	South Africa	Kaapvaal Craton/foreland basin	Mesoarchaean	96 703	2–10
Berelekh	Russia	Siberian Platform	Mesozoic/Recent	2179	12
Tarkwa	Ghana	West African Craton, rift?	Palaeoproterozoic	2158	1.5
Orogenic and intrusion-related deposits, total				74 345	
Muruntau	Uzbekistan	Tien Shan orogen	Permian	6137	3.5
Ashanti	Ghana	West African Craton	Palaeoproterozoic	3169	2–7
Golden Mile	Australia	Yilgarn Craton	Neoarchaean	2079	2
Telfer	Australia	Paterson Orogen	Neoproterozoic	1527	1.5
Homestake	Canada	Trans-Hudson Orogen	Palaeoproterozoic	1237	8.3
Sukhoi Log	Russia	Siberian Craton	Carboniferous	1361	2.5
Volcanic arc-hosted porphyry Cu–Au(–Mo), total				21 240	
Grasberg	Indonesia	Island Arc	Neogene	6817	0.9
Kalmakyrsk	Uzbekistan	Continental Arc	Carboniferous	1299	0.6
Boddington	Australia	Yilgarn Craton	Neoarchaean	1277	0.9
Cananea	Mexico	Continental Arc	Neogene	1269	0.3
Ok Tedi	Papua New Guinea	Island Arc	Neogene	1128	1.0
Porgera	Papua New Guinea	Island Arc	Neogene	1113	3.5
Bingham	USA	Continental Arc	Palaeogene	1001	0.3
Epithermal, total				17 971	
Ladolam (Lihir)	Papua New Guinea	Island Arc	Neogene	2074	3.2
Yanacocha	Peru	Continental Arc	Neogene	1956	0.9
Carlin-type, total				10 013	
Newmont Nevada	USA	Back-arc	Palaeogene	2788	1.7
Betze Post	USA	Back-arc	Palaeogene	1702	4.1
Carlin	USA	Back-arc	Palaeogene	1278	1.6
Cortez	USA	Back-arc	Palaeogene	1258	1.4
Ironoxide-copper–gold deposits, total				2884	
Olympic Dam	Australia	Gawler Craton	Mesoproterozoic	1995	0.5
Au-rich Volcanic-hosted massive sulphide, total		Back-arc		1611	
Skarn, total		Volcanic arc		1467	
Liquid-magmatic, total		Ultramafic complex		1333	
Syn-sedimentary, total		passive margin		1070	

Rudnick and Gao, 2005; Togashi and Terashima, 1997; Yokoyama et al., 1996). Today's volume of crust can be calculated as between 5.8 and 6.9 billion km^3 from the spatial distribution of crust on the Earth's surface and crustal thickness derived from seismic data, modeled eroded thickness and petrological evidence for the lithospheric thickness (Abbott et al., 2000). Assuming an average continental crustal Au content of 1.5 $\mu g\ kg^{-1}$ and a total continental crustal mass of 2.97×10^{19} t (Albarède, 2003), a total of some 45 Gt Au should be present in Earth's continental crust. This is an enormous amount compared to the approximately 183,000 t Au that has been mined to date. The picture does not change significantly if we add the known reserves and resources of approximately 32,000 and 96,000 t, respectively, as obtained from available company reports, thus yielding a total of known Au concentrated in gold deposits of approximately 311,000 t.

If only the top 4 km of the crust are considered viable for mining (currently the deepest mines in the world are gold mines in the Witwatersrand that reach such a depth) and taking the average crustal thickness to be 40 km, all gold known to be concentrated in deposits amounts to not more than 7×10^{-7} of all gold theoretically present in the crust. Such an extremely low proportion could mean that mankind has not been successful at all in locating gold deposits and that huge hitherto unknown deposits still await to be discovered. Alternatively, and more probably, it could mean that geologic processes have been extremely inefficient in concentrating gold into economic ore bodies due to the very low reactivity of gold.

To substantiate the above interpretation, the likely Au solubility in typical geologic transport media may be scrutinized. Most crustal fluids are predominantly aqueous with variable proportion of carbonic species (mainly CO_2). They are of low salinity if derived from meteoric waters and/or prograde metamorphic dehydration reactions, but of high salinity if derived from magmas, retrograde metamorphic hydration reactions or the reaction with evaporite deposits in sedimentary basins. The majority of known gold deposits show evidence of gold precipitation from aqueous-carbonic, sulphur-bearing

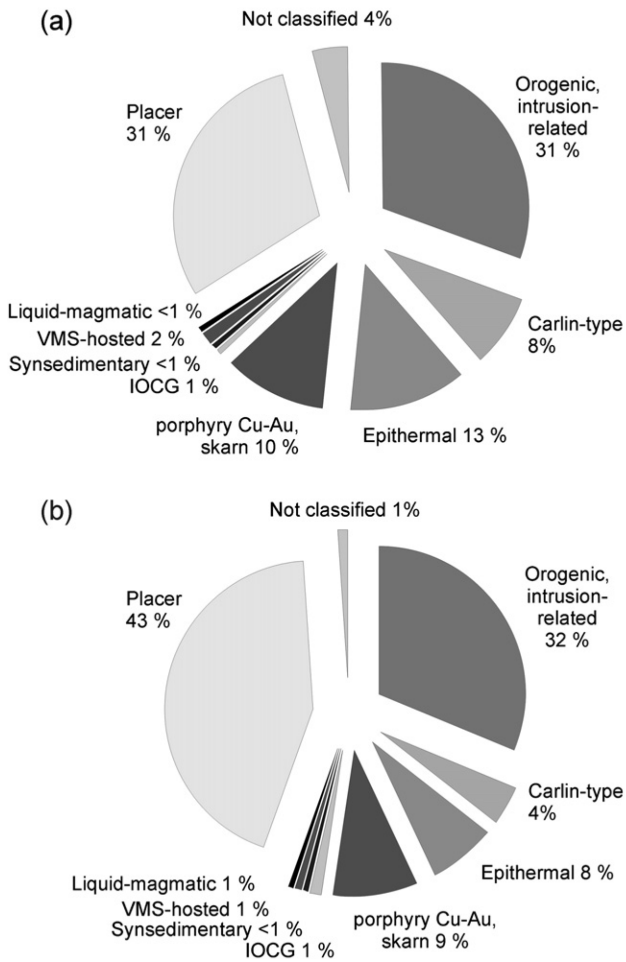

Fig. 2. Relative significance of different gold deposit types in terms of (a) recent global production (1984–2006) and (b) total past production, reserves and resources combined; historic data are taken into account only for those deposits, for which a genetic classification is possible.

hydrothermal fluids (temperature >200 °C) of relatively low salinity and a redox state that is more reducing than the haematite-magnetite buffer (Phillips and Evans, 2004). Under these conditions, gold is carried in solution mainly as AuHS(aq) at low pH and as $Au(HS)^-$ at circum-neutral pH conditions, whereas only at temperatures above 400 °C, AuOH(aq) and $AuCl_2^-$ become important additional complexes, depending on overall sulphide and chloride concentration (Stefansson and Seward, 2004). For the lower temperature range, the latter authors reported measured Au solubilities between 0.007 and 131 mg kg^{-1}, which covers the range of Au concentrations reported from different geothermal fields: the lower end of the range corresponds to Au solubilities in those geothermal systems that are fed by meteoric waters and do not form gold deposits (New Zealand: Pope et al., 2005), whereas the upper range is on a similar order of magnitude as the Au solubility (1–16 mg kg^{-1}) in deep geothermal volcanogenic brines which have formed the giant epithermal Ladolam gold deposit (Papua New Guinea; Simmons and Brown, 2006). Comparable Au-transport capacity is displayed by heated seawater that has infiltrated into, and reacted with, young oceanic crust (Hannington et al., 2005). Hydrothermal fluid flow in volcanic environments may be pervasive or strongly focused — with Au flux on the order of 10^2 to 10^3 kg yr^{-1} (Simmons and Brown, 2006).

Modern meteoric waters and seawater lack the necessary S content to carry significant amounts of Au-hydrosulphide complexes. This is reflected by seawater, in spite of its salinity, having Au concentrations (Falkner and Edmond, 1990) that are five to six orders of magnitude lower than in gold-forming, volcanogenic geothermal brines. Metamorphic fluids, in particular residual fluids after retrograde hydration reactions, have elevated salinity. Depending on lithology, S contents may be elevated due to desulphidation reactions, and CO_2, derived from decarbonatization reactions in the crust or from mantle degassing, may buffer the fluid pH at a level that keeps up elevated Au solubility (Phillips and Evans, 2004). Deep metamorphic fluids are typically relatively reducing and thus can have compositions suitable for the transport of Au-hydrosulphide, or at higher temperatures Au-chloride complexes. Indeed, orogenic gold deposits have been associated with the ascent of such aqueous-carbonic metamorphic fluids (Powell et al., 1991), although a derivation of these fluids from the crystallization of deep-seated granites cannot be ruled out (Ridley and Diamond, 2000). A fundamental problem with the ore-forming potential of metamorphic fluids is the rate of fluid production. Metamorphic rocks typically have very low porosity and permeability with fluid pressure being close to lithostatic. The rate-controlling step for the strongly endothermic dehydration reactions is heat supply, which in turn is very slow considering that regional metamorphism takes place over millions of years. Consequently, the rate of metamorphic fluid production must be correspondingly low. Even if the fluid has elevated Au levels, formation of an ore body would only be possible in the unlikely situation of fluid flow remaining focused along the same structures over millions of years.

In contrast to metamorphic fluids, the amount of formation waters per rock volume is several orders of magnitude greater. These waters tend to be more oxidizing, however, and thus are far less suitable to transport large amounts of gold. Gold deposits that formed directly from the crystallization of a melt are not known and the amount of Au that is produced as by-product of liquid-magmatic deposits is with less than 1% of total Au production negligible here. The inefficiency of silicate melts to carry Au is also indicated by Au concentrations in silicic igneous rocks not exceeding the crustal average (Connors et al., 1993; Ulrich et al., 1999). Most of the Au that is being transported by a melt will be extracted into a fluid phase upon crystallization and degassing of the magma. This is manifest by somewhat lower Au concentrations in some rhyolites (Connors et al., 1993), which is probably related to loss of Au during melt crystallization into the residual fluid. Consequently, magmatic fluids, such as those involved in the formation of porphyry Cu–Au deposits, intrusion-related, IOCG or skarn deposits, volcanogenic epithermal deposits, and possibly even some orogenic gold deposits, can have high Au concentrations. Values of as much as 10^2 and 10^4 mg kg^{-1} have been noted for the vapour and liquid phases, respectively, of porphyry-related Au-ore fluids (Ulrich et al., 1999). In summary, amongst all the

Please cite this article as: Frimmel, H.E., Earth's continental crustal gold endowment, Earth Planet. Sci. Lett. (2007), doi:10.1016/j.epsl.2007.11.022

different fluid types, magmatic fluids seem to have the greatest potential for transporting significant amounts of Au. Compared to metamorphic devolatilization, crystallization and degassing of magma typically take place at much higher rates, which greatly increases the possibility of maintaining focused fluid pathways — an important ingredient for forming an ore body.

4. Gold addition to the continental crust over time

Previous studies on the temporal distribution of gold deposits, in particular orogenic ones (Bierlein et al., 2006; Goldfarb et al., 2001), emphasized the importance of the time around 2.7 Ga for the formation of such deposits ("Global Gold Event"). The amount of gold concentrated into ore bodies at that time is, however, dwarfed by the enormous quantity of gold (almost 97,000 t) in the palaeoplacer deposits of the Witwatersrand Basin. This results in a very strong bias in the age distribution of placer gold deposits in particular and all gold deposits combined (Fig. 3). About 87% of all placer gold is Mesoarchaean, followed by 8%, essentially representing a single unit, the Neoarchaean Ventersdorp Contact Reef above the Witwatersrand Supergroup, and 2.6% of Palaeoproterozoic age. The latter comprises essentially the Tarkwaian deposits in Ghana, with very minor contributions from the Jacobina (genesis ambiguous) and Roraima deposits in Brazil and Guyana. No further significant palaeoplacer gold deposits are known from younger units, except for the Cenozoic placer deposits that make up about 3% of all known placer gold.

The temporal distribution of different types of gold deposit (Fig. 3) clearly shows that the concentration of Au into the various deposit types took place at vastly different rates at different times. These differences are most likely related to the rates of continent formation, evolution of the atmosphere and hydrosphere, and thus to changes in the chemistry of the potential Au-transporting geologic media. The shown distribution is, however, also very strongly influenced by preservation potential.

Primary deposits that form(ed) on or near the surface in volcanic arcs, such as epithermal, porphyry Cu–Au, skarn, or shallow intrusion-related deposits, are known almost exclusively from Cenozoic units. There is no reason to assume that this type of deposit did not form in older, Precambrian volcanic arcs as well, but the probability of preserving such shallow crustal deposits from subsequent erosion is slim. In contrast, known orogenic (including deeper intrusion-related) gold deposits cover a time span of more than 3.4 billion years, but their temporal distribution is punctuated with peaks at 2.7 to 2.5 Ga, 2.1 to 2.0 Ga, 1.9 to 1.7 Ga, 0.8 to 0.6 Ga, 0.45, 0.35, 0.3 to 0.25 Ga, and 0.19 to 0.05 Ga (Goldfarb et al., 2001). As these deposits formed at deeper crustal levels, their preservation potential is considerably higher and consequently, the observed temporal distribution pattern might correspond more closely to the true distribution. A good correlation between timing of orogenic gold deposits and continental crustal growth has been highlighted previously (Groves et al., 2005), but it fails to explain the by far largest known concentration of Au in Archaean palaeoplacer deposits.

To get a better idea of Au productivity over geologic time, the amounts of gold in different deposit types as shown in Fig. 3 were related to the proportion of exposed areas underlain by rocks of the various aeras and normalized to time slices of 100 million years (Fig. 4). Historical (pre-19th century) gold for which no production figures are available, such as gold from the Arabian–Nubian Shield or the European Variscan units, was excluded from this analysis, but based on previous rough estimates for that gold (Goldfarb et al., 2001), this is taken to be negligible in the larger picture of interest here. Fig. 4 shows an even more pronounced bimodal distribution with extremely high Au productivity in the Archaean and apparently again in the Cenozoic. The latter peak reflects the widespread occurrence of young Cenozoic mountain belts that have not been eroded yet. About 33% of the land surface is underlain by Cenozoic rocks (calculated from data in Chorlton, 2007). As most of the Cenozoic gold is located at very shallow crustal levels (Fig. 3), its preservation potential in older tectonic units, as pointed out above, would be very low. Thus a higher proportion of younger gold deposits is predicted. Less easily predicted is the enormous bias towards Archaean gold, considering that the bulk of that gold is present as palaeoplacer deposits from a single gold province, the Witwatersrand.

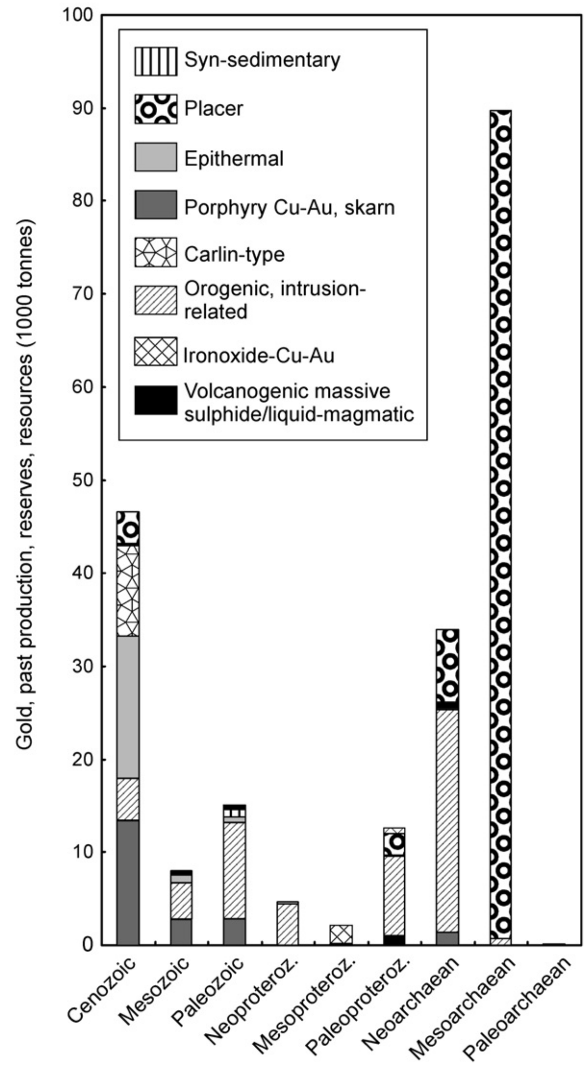

Fig. 3. Distribution of known gold deposit types over geologic time.

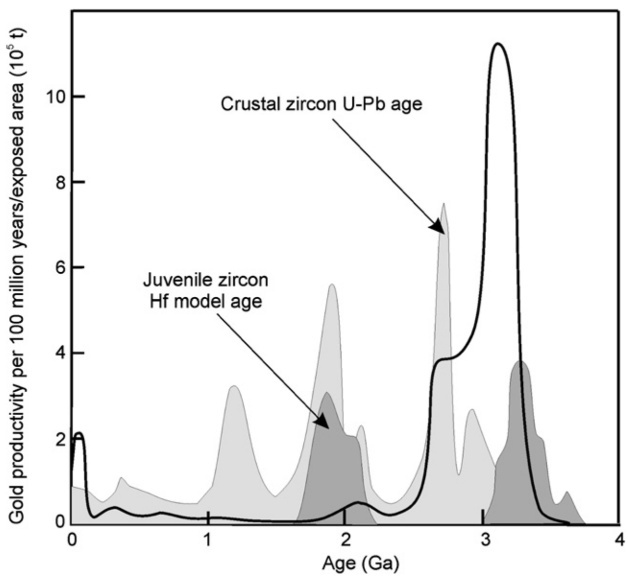

Fig. 4. Probability distribution of gold productivity (in 10^5 t) per 100 million years normalized to the relative exposed areas underlain by rocks of different aeras (calculated from areas as given in Chorlton, 2007). Also shown are crustal zircon age distributions (Condie, 1998) and Hf model age distributions obtained on zircon from juvenile crust (Kemp et al., 2006).

The same argument as for the shallow crustal gold deposits above should apply even more to placer deposits, bearing in mind that they typically form on the surface and in environments that are particularly prone to subsequent erosion. Yet, more than 90% of all placer gold stems from the Archaean, with only 5.7% from the Cenozoic. A certain amount of Cenozoic placer gold produced by informal small-scale miners is likely to have been omitted from official production records, but this uncertainty has little bearing on the fact that the overwhelming majority of gold in general, and of placer gold in particular, has come from the Witwatersrand. This supergiant amongst gold provinces has been explained by a combination of factors. These include the presence of an Au-enriched hinterland (i.e. Palaeo- to Mesoarchaean granite-greenstone belts), a foreland basin depositional setting that, combined with Archaean environmental conditions (no vegetation cover on land, intense chemical weathering), made possible particularly vigorous sediment-reworking to occur, and an exceptional preservation beneath a thick cover of Neoarchaean flood basalt and in the middle of one of the oldest and most stable cratons (Frimmel et al., 2005). Other Archaean placer deposits comparable to those of the Witwatersrand might have existed also on other Archaean continents but would have been eroded or recycled on a lithosphere-scale during subsequent plate tectonic processes. Consequently, the proportion of gold having been added to the continental crust should have been even greater in the Archaean than is reflected in the already strongly skewed distribution of known gold towards Archaean deposits as shown in Fig. 4.

Thus the question arises as to reason(s) for the extraordinarily high rates of Au sequestration from whatever source into the continental crust in the Mesoarchaean, which must have been orders of magnitude higher than later in Earth's history. It cannot be excluded that such high (or even higher) Au sequestration rates existed already in the Palaeoarchaean (and before), but are not recorded because of the lack of preservation of suitable rock sequences from those early times. A fundamental clue to this question is provided by the gold chemistry. The Witwatersrand gold is compositionally very different to all younger gold studied to date: several orders of magnitude more Re and Os have been reported (Frimmel et al., 2005; Kirk et al., 2001, 2002) from Witwatersrand gold compared to younger, orogenic or epithermal gold (Fig. 5). The exceptionally high Os concentrations between 2 and 10350 ppm noted for Witwatersrand gold are incompatible with gold precipitation from aqueous hydrothermal fluids, because of the generally very low solubility of Os in such fluids (Xiong and Wood, 2000). This effectively rules out Au precipitation from a post-depositional hydrothermal fluid as suggested by those who have advocated a hydrothermal model for the Witwatersrand gold, but it also argues against derivation of detrital gold particles from the erosion of orogenic, hydrothermal gold deposits in the Meso- to Palaeoarchaean greenstone belts in the hinterland, as suggested by many workers who prefer a palaeoplacer model (e.g. Robb and Meyer, 1990).

The high Re and in particular Os concentrations in the Witwatersrand gold are most compatible with derivation from magmatic sources, more specifically from rocks that formed by high degrees of partial melting in the mantle. Higher degrees of partial melting in the Archaean mantle are expected as the overall heat flux was most likely greater in the young Earth because of a higher radiogenic heat production (Pollack, 1997). Alternatively, more voluminous melt production has been explained by a wetter mantle (Parman et al., 1997). Gold derivation from such a mantle is supported by the Re/Os ratio in the Witwatersrand gold, which is close to that of the mantle, and the initial $^{187}Os/^{188}Os$ ratio of 0.108, which corresponds to that estimated for the mantle at about 3.0 Ga (Kirk et al., 2002).

So far, Au concentrations reported from Archaean greenstone belts do not exceed significantly the crustal average (Meyer and

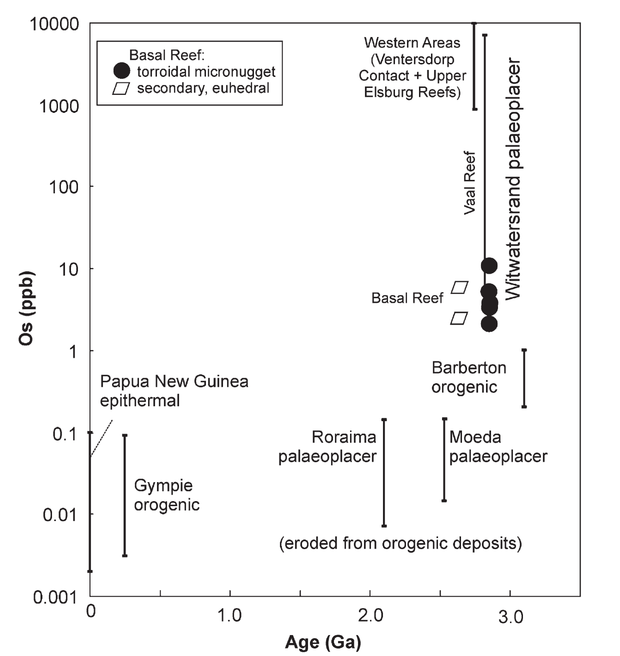

Fig. 5. Ranges of Os concentrations in gold of different age (data from Kirk et al., 2002, and Frimmel et al., 2005).

Please cite this article as: Frimmel, H.E., Earth's continental crustal gold endowment, Earth Planet. Sci. Lett. (2007), doi:10.1016/j.epsl.2007.11.022

Saager, 1985; Stone and Crocket, 2003). A much more effective gold extraction mechanism from whatever source rocks is therefore called for. The widespread occurrence of detrital pyrite in fluvial deposits attests to elevated H_2S fugacity and reducing conditions in Archaean meteoric environments (Frimmel, 2005; Krupp et al., 1994). Although the Au solubility under the inferred low pH, estimated at approximately 4 (Krupp et al., 1994), decreases (Stefansson and Seward, 2004), the net effect of the higher H_2S and extremely low O_2 fugacities would have been an Au solubility that is up to four orders of magnitude higher. This, combined with deep chemical weathering, as indicated for the Archaean (Frimmel, 2005), and high erosion rates, helped by the lack of land vegetation, would have led to a markedly higher fluvial Au run-off from the continent. In contrast to the pyrite-rich fluvial to fluvio-deltaic deposits, magnetite is the principal Fe-phase in distal marine sediments of the Witwatersrand, implying several orders of magnitude lower total sulphur concentrations in the contemporary seawater (Frimmel, 2005). Under such conditions the Au solubility drops rapidly, which might explain why Archaean shales and banded iron formation contain considerable more Au than younger marine shales (on average 70 μg kg^{-1}, Meyer and Saager, 1985).

A more fertile hinterland in Mesoarchaean times cannot be excluded, however. What is preserved today as granitoid–greenstone belts in the Kaapvaal Craton are the eroded remnants of potential source areas. As established above, convergent plate margins are by far the best setting for large gold concentrations — disregarding placer deposits they host 87% of all known gold. By analogy it is speculated that the bulk of the Archaean gold was added to the crust in a similar way, i.e. within ancient volcanic arcs. Calcalkaline magmatic rocks with geochemical and isotopic signatures typical of a supra-subduction setting occur in the immediate basement of the Witwatersrand Basin and correlatives (Lehrmann and Frimmel, 2007; Wilson and Grant, 2006), and these may be evidence of the existence of such a volcanic arc as source domain. Note that elsewhere, the third largest known porphyry-style gold deposit, Boddington (Table 1), is Neoarchaean in age and thus shows that this style of Au-mineralization has been already in operation in Archaean times.

It has been established above that crustal-scale gold mobilization is best achieved by aqueous-carbonic fluids and not by melts. Every time Os-rich gold becomes remobilized by an aqueous fluid, its Os content should decrease because of the comparatively very low Os solubility. This effect should be pronounced particularly strongly in the case of a fluid-buffered system and it is well reflected by the very low Os concentrations in gold of undoubtedly hydrothermal origin, such as palaeoplacer gold in the Roraima and Moeda deposits of Guyana and Brazil, respectively, both derived from Palaeoproterozoic orogenic gold deposits, the Permotriassic orogenic gold from Gympie (eastern Australia) and the Holocene epithermal gold from Papua New Guinea (Fig. 5). Gold from these deposits is depleted in Os by several orders of magnitude. The opposite behaviour is noted in the two generations of gold that have been described from the Basal Reef in the Witwatersrand (Minter et al., 1993). There, well rounded, pitted, torroidal to disk-shaped, c. 3.0 Ga detrital gold micronuggets occur together with clearly secondary, euhedral gold crystals or irregularly shaped, dendritic gold of undoubtedly hydrothermal nature on a micro- to millimeter scale. The co-existence of the two gold generations on such a small scale provides perfect evidence of very short-range mobilization of detrital gold particles within the host conglomerate. Both generations have almost identical Os concentrations (Fig. 4), highlighting the mineral-buffered, local, and only very limited fluid-rock interaction in that particular unit.

Bearing in mind that the Cenozoic peak in the age distribution of known gold is a result of much greater preservation of younger rocks, the actual proportion of Archaean gold must be much higher than shown in Fig. 3 (58%), possibly even more than 82% — the proportion shown in Fig. 4, with an almost exponential decrease in gold addition to the continental crust since the Archaean. Mesoarchaean orogenic gold from the Barberton greenstone belt takes an intermediate position between the Witwatersrand gold and the younger, hydrothermal gold examples in terms of Os content and Re/Os ratio (Frimmel et al., 2005). This is explained by that Mesoarchaean gold not having been re-mobilized by crustal fluids as often as younger hydrothermal gold. Overall, the decrease in Os concentration in the gold over time, combined with the observation of the majority of known gold being Archaean, may be explained by the bulk of the gold having been added to the continental crust already by the Mesoarchaean and since then having been recycled repeatedly by various crustal fluids.

Large uncertainties exist in the determination of continental growth rates over time. One end-member model assumes that continental crust grew continuously (though in episodic pulses) and largely irreversibly over the past four billion years (e.g., Fyfe, 1983; McLennan and Taylor, 1982). The other assumes that the mass of continental crust remained more or less constant for the past four billion years with crust recycling into the mantle having taken place at the same rate as crust formation (Armstrong, 1991). Continental crust formation as early as 4.4 to 4.5 Ga is now convincingly indicated by Hf isotopes in detrital zircon grains (Harrison et al., 2005). Subsequent further continental growth is assumed by most workers to have been episodic with peaks at 3.0, 2.7, 2.1 to 1.9 and 1.2 Ga (Abbott et al., 2000; Condie, 1998; Green et al., 2000; Parman, 2007). Much of that growth involved the recycling of older crust, but strong evidence exists for juvenile crust addition in the periods 3.6 to 3.0 Ga (peak at 3.2–3.1 Ga) and 2.1 to 1.9 Ga, based on zircon grains with low $\delta^{18}O$ (indicative of crystallisation from melts with negligible sedimentary component) and Hf model ages (Kemp et al., 2006). These two peaks in juvenile crust formation correspond with the bulk of known gold ages, i.e. 3.3 to 3.0 Ga (58% of all known gold), most of it referring to the source of the Witwatersrand gold, and 2.1 to 1.9 Ga (6%), mainly orogenic and derived placer gold from the Birimian and related belts in West Africa and northwestern South America (Fig. 4).

The scarcity of Palaeoarchaen, and effective lack of Hadaean, gold deposits makes it impossible to ascertain whether the apparent peak in the addition of gold to the continental crust in the Mesoarchaean, as is seemingly inferred from Figs. 3 and 4, is a true reflection of the gold sequestration rate over time. The lack of older than Mesoarchaean gold deposits might reflect a very high degree of crustal recycling during the Eo- to Palaeoarchaean.

Alternatively, the extraordinary amount of Mesoarchaean gold may be explained by a period of particularly strong meteorite bombardment. With a Au content of about 140 μg/kg in chondrite (McDonough and Sun, 1995), undifferentiated planetesimals contain two orders of magnitude more Au than the Earth's mantle and crust. Addition of such extraterrestrial material to the Earth after core-mantle differentiation would have led to an increase in the Au content of the mantle and crust. A period of particularly heavy meteorite bombardment during the Archaean has been suggested, based on age distribution patterns for lunar impact structures and mare basalt for the time at approximately 3.2 Ga (Glikson, 2001), which is close to the apparent peak in the age distribution of known gold.

A further alternative, which is preferred here, assumes an extraordinary Mesoarchaean gold event that is a consequence of the thermal history of the mantle. In a recent study involving the modelling of the mantle's thermal evolution from compositional variations of mantle melts over time and from rheological constraints for the beginning of sub-solidus convection at the end of the initial magma ocean phase (Labrosse and Jaupart, 2007) it has been concluded that the mantle temperature reached a maximum at about 3 Ga (250° hotter than today and about 100° hotter than 4 Ga ago). Interestingly, this calculated thermal peak is indepent of the assumed continental growth rate. Considering the principle gold reservoir in Earth being in the core, mantle plumes provide a most effective transfer mechanism for gold from the core-mantle boundary to the lithosphere. It has been speculated, for instance, that the Yellowstone hotspot might have caused the Eocene Carlin-type gold province in Nevada (Oppliger et al., 1997). During Eo- to Palaeoarchaean times, plume activity was most likely far more vigorous. A thermal peak in the mantle at around 3 Ga would thus provide an explanation not only for the change from dominantly plume- to plate tectonics, but it would also explain the principal sequestration of gold from a postulated hot mantle into the continental crust at that time.

The formation of juvenile crust has been recognized as a first-order control on the likelihood of developing giant orogenic gold systems based on the global distribution of such deposits (Bierlein et al., 2006), whereby hydrated mafic crust is interpreted as the best available source of both the mineralizing fluids and the gold. Giant orogenic gold provinces seem to occur preferentially in orogens with subducted oceanic crust or only thin continental lithosphere, where the pre-mineralization crustal history had been short. The significance of oceanic crust as potential gold source for orogenic gold deposits is highlighted by the Au-enrichment of hydrothermal fluids on the ocean floor (Hannington et al., 2005). An apparent exception to the postulated relationship between giant orogenic gold deposits and lithospheric thinning, expressed by the short duration of pre-mineralization crustal history, is the giant Muruntau (>6100 t Au) and related deposits in the Tien Shan orogen. There mineralization was coeval with a Variscan orogenic event that followed an earlier Caledonian orogeny, thus reflecting a long pre-mineralization crustal history. Relatively unradiogenic initial Os isotope ratios and elevated $^3He/^4He$ ratios (Graupner et al., 2006; Morelli et al., 2007) indicate a mantle-derived component in the ore fluid there, thus pointing to a significant phase of extension, which made it possible for post-orogenic melts to ascend and for mantle fluids to contribute to the ore fluid at mid-crustal levels. This is an important finding, because it shows that even in a thick lithospheric setting, some of the mineralizing fluid is derived from the underlying mantle. A mantle source for the fluid does not necessarily imply a mantle source for the gold too. However, while much of the gold in post-Archaean continental crust has been recycled, as the crustal material itself, a small proportion of that gold may be juvenile. The Tien Shan gold province may well be the best example of such juvenile, mantle-derived post-Archaean gold. This would explain not only the noted difference from other orogenic gold provinces with regard to tectonic setting and its strong association with post-orogenic 285–290 Ma intrusive activity as documented by radiometric age data (Mao et al., 2004; Morelli et al., 2007). It would also explain why the deposits in the Tien Shan are much more endowed with gold than any other known orogenic gold deposit.

5. Conclusions

The analysis of past production data as well as reserve and resource estimates for the principal genetic gold deposit types of different age leads to the conclusion that the Witwatersrand palaeoplacer and orogenic gold deposits are with 40 and 32% of known gold, respectively, by far the two economically most important types of gold deposits. The Cenozoic and Mesoarchaean Aeras are apparently the most important times for the formation of gold deposits (12 and 82% of known global gold, respectively). As the large proportion of Cenozoic deposits is an artifact of the much higher preservation potential of younger deposits, the actual temporal distribution of gold deposits is strongly biased towards the Archaean. Most of the crustal gold was sequestered from the mantle already by the Mesoarchaean, probably by H_2O-rich melts as typically found in active plate margins. Subsequently, that gold was recycled repeatedly on a lithospheric scale by a variety of crustal fluids, predominantly of magmatic origin. Remarkably variable but overall very high Os concentrations and unradiogenic Os isotope ratios in the Witwatersrand gold highlight its relatively juvenile character with only some remobilization by crustal fluids prior to deposition as placer deposits. In contrast, most of the younger gold has been recycled extensively, thus loosing most of its original Os. An example of post-Archaean mineralizing fluids that emanated from the mantle may be found in the Tien Shan gold province, notably the giant Muruntau deposit. It remains to be tested, whether other giant orogenic or intrusion-related gold deposits also contain a mantle component. Even if they did, it would not change the fact that the overwhelming part of all continental crustal gold was extracted from the mantle as early as in the Archaean.

The actual cause for the very high rates of gold addition to the continental crust in Archaean times remains open to debate. It may be a question of juvenile crust formation rate — in itself a controversial issue. In that case, the conclusion reached here would support the model of most of the continental crust having formed already in the Archaean or earlier. While the possibility of an extraordinary Mesoarchaean gold-enrichment as a consequence of major meteorite bombardment at approximately

3.2 Ga (Glikson, 2001) cannot be dismissed entirely, it seems more likely that the Mesoarchaean gold event resulted from an extraordinary high gold flux from the mantle into the crust during the transition from a plume world to a plate-tectonic world when the mantle temperature reached its maximum.

The overall proportion of gold that became concentrated into known ore bodies over time is very small and is less than the proportion of gold that is dissolved in modern crustal fluids. This holds promise for a large proportion of gold deposits still remaining to be discovered.

Acknowledgements

Two anonymous reviewers are thanked for helpful comments as well as C.P. Jaupart for editorial handling. Financial support from the Deutsche Forschungsgemeinschaft (grant FR2183/3-1) is gratefully acknowledged. This is a contribution to the International Geoscience Programme IGCP 540.

References

Abbott, D., Sparks, D., Herzberg, C., Mooney, W., Nikishin, A., Zhang, Y.S., 2000. Quantifying Precambrian crustal extraction: the root is the answer. Tectonophysics 322, 163–190.

Albarède, F., 2003. Geochemistry, An Introduction. Cambridge University Press, Cambridge. 248 pp.

Armstrong, R.L., 1991. The persistent myth of crustal growth. Aust. J. Earth Sci. 38, 613–630.

Bierlein, F.P., Groves, D.I., Goldfarb, R.J., Dube, B., 2006. Lithospheric controls on the formation of provinces hosting giant orogenic gold deposits. Miner. Depos. 40, 874–886.

Chorlton, L., 2007. Generalized geology of the world: bedrock domains and major faults in GIS format. Geological Survey of Canada, Open File 5529, Ottawa. 1 CD-ROM.

Cline, J.S., Hofstra, A., Muntean, J.L., Tosdal, R.M., Hickey, K.A., 2005. Carlin-type gold deposits in Nevada: critical geologic characteristics and viable models. In: Hedenquist, J.W., Thompson, J.F.H., Goldfarb, R.J., Richards, J.P. (Eds.), Economic Geology One Hundreth Anniversary Volume. Society of Economic Geologists, Littleton, pp. 451–484.

Condie, K.C., 1998. Episodic continental growth and supercontinents: a mantle avalanche connection? Earth Planet. Sci. Lett. 163, 97–108.

Connors, K.A., Noble, D.C., Bussey, S.D., Weiss, S.I., 1993. Initial gold contents of silicic volcanic rocks: bearing on the behavior of gold in magmatic systems. Geology 21, 937–940.

Emsbo, P., Hofstra, A.H., Lauha, E.A., Griffin, G.L., Hutchinson, R.W., 2003. Origin of high-grade gold ore, source of ore fluid components, and genesis of the Meikle and neighboring Carlin-type deposits, northern Carlin trend, Nevada. Econ. Geol. 98, 1069–1100.

Emsbo, P., Koenig, A.E., 2007. Transport of Au in petroleum: evidence from the northern Carlin trend, Nevada. In: Andrew, C.J., et al. (Eds.), Digging Deeper, Proceedings of the 9th Biennial SGA Meeting, 20–23 August 2007, vol. 1. Irish Association for Economic Geology, Dublin, pp. 695–698.

Falkner, K.K., Edmond, J.M., 1990. Gold in seawater. Earth. Planet. Sci. Lett. 98, 208–221.

Frimmel, H.E., 2005. Archaean atmospheric evolution: evidence from the Witwatersrand gold fields, South Africa. Earth-Sci. Rev. 70, 1–46.

Frimmel, H.E., Groves, D.I., Kirk, J., Ruiz, J., Chesley, J., Minter, W.E.L., 2005. The formation and preservation of the Witwatersrand goldfields, the largest gold province in the world. In: Hedenquist, J.W., Thompson, J.F.H., Goldfarb, R.J., Richards, J.P. (Eds.), Economic Geology One Hundreth Anniversary Volume. Society of Economic Geologists, Littleton, pp. 769–797.

Fyfe, W.S., 1983. The evolution of Earth's crust: modern plate tectonics to ancient hot spot tectonics? Chem. Geol. 23, 89–114.

Glikson, A.Y., 2001. The astronomical connection of terrestrial evolution: crustal effects of post-3.8 Ga mega-impact clusters and evidence for major 3.2±0.1 Ga bombardment of the Earth–Moon system. J. Geodyn. 32, 205–229.

Goldfarb, R.J., Baker, T., Dube, B., Groves, D.I., Hart, C.J.R., Gosselin, P., 2005. Distribution, character, and genesis of gold deposits in metamorphic terranes. In: Hedenquist, J.W., Thompson, J.F.H., Goldfarb, R.J., Richards, J.P. (Eds.), Economic Geology One Hundreth Anniversary Volume. Society of Economic Geologists, Littleton, pp. 407–450.

Goldfarb, R.J., Groves, D.I., Gardoll, S., 2001. Orogenic gold and geologic time: a global synthesis. Ore Geol. Rev. 18, 1–75.

Gosselin, P., Dube, B., 2005. Gold deposits of the world: distribution, geological parameters and gold content. Geological Survey of Canada, Open File 4895, Ottawa, p. 221.

Graupner, T., Niedermann, S., Kempe, U., Klemd, R., Bechtel, A., 2006. Origin of ore fluids in the Muruntau gold system: constraints from noble gas, carbon isotope and halogen data. Geochim. Cosmochim. Acta 70, 5356–5370.

Green, M.G., Sylvester, P.J., Buick, R., 2000. Growth and recycling of early Archaean continental crust: geochemical evidence from the Coonterunah and Warrawoona Groups, Pilbara Craton, Australia. Tectonophysics 322, 69–88.

Groves, D.I., Condie, K.C., Goldfarb, R.J., Hronsky, J.M.A., Vielreicher, R.M., 2005. Secular changes in global tectonic processes and their influence on the temporal distribution of gold-bearing mineral deposits. Econ. Geol. 100, 203–224.

Handley, J.R.F., 2004. Historic Overview of the Witwatersrand Goldfields. Handley, Howick. 224 pp.

Hannington, M.D., de Ronde, C.E.J., Petersen, S., 2005. Sea-floor tectonics and submarine hydrothermal systems. In: Hedenquist, J.W., Thompson, J.F.H., Goldfarb, R.J., Richards, J.P. (Eds.), Economic Geology One Hundreth Anniversary Volume. Society of Economic Geologists, Littleton, pp. 111–141.

Hannington, M.D., Poulsen, K.H., Thompson, J.F.H., Sillitoe, R.H., 1999. Volcanogenic gold in the massive sulfide environment. Rev. Econ. Geol. 8, 325–356.

Harrison, T.M., Blichert-Toft, J., Müller, W., Albarede, F., Holden, P., Mojzsis, S.J., 2005. Heterogeneous Hadean hafnium: evidence of continental crust at 4.4 to 4.5 Ga. Science 310, 1947–1950.

Henry, C.D., Ressel, M.W., 2000. Eocene magmatism of northeastern Nevada: the smoking gun for Carlin-type gold deposits. In: Cluer, J.K., Price, J.G., Struhsacker, E.M., Hardyman, R.F., Morris, C.L. (Eds.), Geology and Ore Deposits 2000: The Great Basin and Beyond. Geological Society of Nevada, Reno, pp. 365–388.

Hofstra, A., Cline, J.S., 2000. Characteristics and models for Carlin-type gold deposits. Rev. Econ. Geol. 13, 163–220.

Kemp, A.I.S., Hawkesworth, C.J., Paterson, B.A., Kinny, P.D., 2006. Episodic growth of the Gondwana supercontinent from hafnium and oxygen isotopes in zircon. Nature 439, 580–583.

Kirk, J., Ruiz, J., Chesley, J., Titley, S., Walshe, J., 2001. A detrital model for the origin of gold and sulfides in the Witwatersrand basin based on Re–Os isotopes. Geochim. Cosmochim. Acta 65, 2149–2159.

Kirk, J., Ruiz, J., Chesley, J., Walshe, J., England, G., 2002. A major Archean gold and crust-forming event in the Kaapvaal Craton, South Africa. Science 297, 1856–1858.

Krupp, R., Oberthür, T., Hirdes, W., 1994. The Early Precambrian atmosphere and hydrosphere: thermodynamic constraints from mineral deposits. Econ. Geol. 89, 1581–1598.

Labrosse, S., Jaupart, C., 2007. Thermal evolution of the Earth: secular changes and fluctuations of plate characteristics. Earth Planet. Sci. Lett. 260, 465–481.

Lehmann, B., Nägler, T.F., Holland, H.D., Wille, M., Mao, J., Pan, J., Ma, D., Dulski, P., 2007. Highly metalliferous carbonaceous shale and Early Cambrian seawater. Geology 35, 403–406.

Lehrmann, B., Frimmel, H.E., 2007. Mesoarchaean basement of the Witwatersrand: a possible source of the gold? In: Andrew, C.J., et al. (Eds.), Digging Deeper, Proceedings of the 9th Biennial SGA Meeting, 20–23 August 2007, vol. 1. Irish Association for Economic Geology, Dublin, pp. 15–18.

Mao, J., Konopelko, D., Seltmann, R., Lehmann, B., Chen, W., Wang, Y., Eklund, O., Usubaliev, T., 2004. Postcollisional age of the Kumtor gold deposit and timing of Hercynian events in the Tien Shan, Kyrgyzstan. Econ. Geol. 99, 1771–1780.

McDonough, W.F., Sun, S.-S., 1995. The composition of the Earth. Chem. Geol. 120, 223–253.

McLennan, S.M., Taylor, R.S., 1982. Geochemical constraints on the growth of the continental crust. J. Geol. 90, 347–361.

Meyer, M., Saager, R., 1985. The gold content of some Archaean rocks and their possible relationship to epigenetic gold-quartz vein deposits. Miner. Depos. 20, 284–289.

Minter, W.E.L., Goedhart, M.L., Knight, J., Frimmel, H.E., 1993. Morphology of Witwatersrand gold grains from the Basal Reef: evidence for their detrital origin. Econ. Geol. 88, 237–248.

Morelli, R., Creaser, R.A., Seltmann, R., Stuart, F.M., Selby, D., Graupner, T., 2007. Age and source constraints for the giant Muruntau gold deposit, Uzbekistan, from coupled Re–Os–He isotopes in arsenopyrite. Geology 35, 795–798.

Muntean, J.L., Frimmel, H.E., Phillips, N., Law, J., Myers, R., 2005. Controversies on the origin of world-class gold deposits, part II: Witwatersrand gold deposits. SEG Newsletter 60, 7–19.

Oppliger, G.L., Murphy, B.J., Brimhall, G.H., 1997. Is the ancestral Yellowstone hotspot responsible for the Tertiary "Carlin" mineralization in the Great Basin of Nevada? Geology 25, 627–630.

Palme, H., O'Neill, H.S.C., 2005. Cosmochemical estimates of mantle composition. In: Carlson, R.W. (Ed.), The Mantle and Core. Treatise on Geochemistry. Elsevier, Amsterdam, pp. 1–38.

Parman, S.W., 2007. Helium isotopic evidence for episodic mantle melting and crustal growth. Nature 446, 900–903.

Parman, S.W., Dann, J.C., Grove, T.L., de Wit, M.J., 1997. Emplacement conditions of komatiite magmas from the 3.49 Ga Komati formation, Barberton Greenstone belt, South Africa. Earth Planet. Sci. Lett. 150, 303–323.

Phillips, G.N., Evans, K.A., 2004. Role of CO_2 in the formation of gold deposits. Nature 429, 860–863.

Pollack, H.N., 1997. Thermal characteristics of the Archaean. In: de Wit, M.J., Ashwal, M.D. (Eds.), Greenstone Belts. Clarendon Press, Oxford, pp. 223–232.

Pope, J.G., Brown, K.L., McConchie, D.M., 2005. Gold concentrations in springs at Waiotapu, New Zealand: implications for precious metal deposition in geothermal systems. Econ. Geol. 100, 677–687.

Porter, K.E., Amey, E.B., 2003. Gold Statistics. United States Geological Survey.

Powell, R., Will, T.M., Phillips, G.N., 1991. Metamorphism in Archaean greenstone belts: calculated fluid compositions and implications for gold mineralization. J. metamorph. Geol. 9, 141–150.

Ridley, J.R., Diamond, L.W., 2000. Fluid chemistry of orogenic lode gold deposits and implications for genetic models. In: Hagemann, S.G., Brown, P.E. (Eds.), Gold in 2000. Reviews in Economic Geology. Society of Economic Geologists, Littleton, pp. 141–162.

Robb, L.J., Meyer, F.M., 1990. The nature of the Witwatersrand hinterland: conjectures on the source-area problem. Econ. Geol. 85, 511–536.

Rudnick, R.L., Gao, S., 2005. Composition of the continental crust. In: Rudnick, R.L. (Ed.), The Crust. Treatise on Geochemistry. Elsevier, Amsterdam, pp. 1–64.

Simmons, S.F., Brown, K.L., 2006. Gold in magmatic hydrothermal solutions and the rapid formation of a giant ore deposit. Science 314, 288–291.

Stefansson, A., Seward, T.M., 2004. Gold (I) complexing in aqueous sulphide solutions to 500 °C at 500 bar. Geochim. Cosmochim. Acta 68, 4121–4143.

Stone, W.E., Crocket, J.H., 2003. Platinum-group element contents of chromites from mafic-ultramafic layered flows, Abitibi greenstone belt, Ontario: implications for geochemical fractionation and mineral exploration. Mineral. Petrol. 78, 139–147.

Togashi, S., Terashima, S., 1997. The behavior of gold in unaltered island arc tholeiitic rocks from Izu-Oshima, Fuji, and Osoreyama volcanic areas, Japan. Geochim. Cosmochim. Acta 61, 543–554.

Ulrich, T., Günther, D., Heinrich, C.A., 1999. Gold concentrations of magmatic brines and the metal budget of porphyry copper deposits. Nature 399, 676–679.

Williams-Jones, A.E., Migdisov, A.A., 2007. The solubility of gold in crude oil: implications for ore genesis. In: Andrew, C.J., et al. (Eds.), Digging Deeper, Proceedings of the 9th Biennial SGA Meeting, 20–23 August 2007, vol. 1. Irish Association for Economic Geology, Dublin, pp. 765–768.

Williams, P.J., Barton, M.D., Johnson, D.A., Fontbote, L., De Haller, A., Mark, G., Oliver, N.H.S., Marschik, R., 2005. Iron oxide copper-gold deposits: geology, space-time distribution, and possible models of origin. In: Hedenquist, J.W., Thompson, J.F.H., Goldfarb, R.J., Richards, J.P. (Eds.), Economic Geology One Hundreth Anniversary Volume. Society of Economic Geologists, Littleton, pp. 371–405.

Wilson, A.H., Grant, C.E., 2006. Physical volcanology and compositions of the basaltic lavas in the Archean Nzuse Group, white Mfolozi inlier, South Africa. Geological Society of America, Special Paper 405, pp. 255–289.

Wood, B.J., Walter, M.J., Wade, J., 2006. Accretion of the Earth and segregation of its core. Nature 441, 825–833.

Xiong, Y., Wood, S.A., 2000. Experimental quantification of hydrothermal solubility of platinum-group elements with special reference to porphyry copper environments. Mineral. Petrol. 68, 1–28.

Yokoyama, T., Yokota, T., Hayashi, S., Izawa, E., 1996. Determination of trace gold in rock samples by a combination of two-stage solvent extraction and graphite furnace atomic absorption spectrometry: the problem of iron interference and its solution. Geochem. J. 30, 175–181.

Please cite this article as: Frimmel, H.E., Earth's continental crustal gold endowment, Earth Planet. Sci. Lett. (2007), doi:10.1016/j.epsl.2007.11.022