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Secondary gold structures: Relics of past biogeochemical transformations and implications for colloidal gold dispersion in subtropical environments

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ABSTRACT

Biogeochemical processes are known to drive the cycling of gold via dissolution/re-precipitation reactions that result in the transformation of gold grain in near-surface environments. In this study, placer gold grains were collected from West Coast Creek, Queensland, Australia and characterized using high-resolution electron microscopy. The outer surface of grains contains 98.1 mol% Au and 1.9 mol% Ag. Crevices occur on the surface of grains and are filled with organics and clay minerals in which nanometer-size gold colloids and micrometer-sized octahedral gold platelets are embedded. The formation of these secondary gold structures is attributed to gold precipitation by the availability of reducing agents such as microbiota, residual organics and clays. Bacterioform gold contains 74.7 mol% Au and 25.3 mol% Ag and is also embedded in clay minerals within crevices. From sonicated gold grains, grain surfaces beneath the clay minerals are striated and contain 68.5 mol% Au and 31.5 mol% Ag. This data suggests that dissolution processes occur at the gold grain interface. Varying sizes of gold colloids on the grains suggest that five 'episodes' of gold dissolution/re-precipitation processes occurred; each episode was estimated to be 7.64 ± 4.1 years. Therefore, these grains represent 17.9–58.5 years of gold cycling and mobilization within this subtropical environment. Furthermore, laboratory experiments involving colloidal gold dispersion demonstrated that iron-oxides and organic material from West Coast Creek sediment adsorbed 94.5% of suspended gold colloids. In conclusion, this study highlights the value of nanophase gold characterization for the interpretation of biogeochemical processes affecting gold grain transformation and mobility in near-surface environments. Importantly, this study is the first to estimate the kinetics of biogeochemical gold cycling with regards to colloidal gold dispersion and re-concentration.

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1. Introduction

Placer gold was thought to be derived purely from the weathering of hypogene sources (Liversidge, 1897). However, placer grains are considered to be comprised of gold-silver alloys and pure gold which represent the hypogene source and secondary gold transformation in supergene environments, respectively (Hough et al., 2009; Reith et al., 2010). Recent research has shown that it is also possible that placer grains could be completely comprised of secondary gold nanoparticles (Shuster and Southam, 2014). Physical factors such as flow rate and bed load within fluvial environments contribute to the mechanical reshaping of grains while gold dissolution and re-precipitation also

contribute to grain morphologies (Bowell, 1992; Colin et al., 1989, 1997, 1993; Craw and MacKenzie, 2010; Greffie et al., 1996; Groen et al., 1990; Youngson and Craw, 1993). In addition, the mobility of soluble gold as gold chloride, cyanide and thiosulfate complexes as well as nanoparticles has been described in supergene enrichment systems (Boyle, 1979; Hough et al., 2008; Mann, 1984; Ta et al., 2014; Webster, 1986). Recent research has shown that gold mobilization, precipitation and gold grain formation can be catalyzed by the biosphere especially by bacterial biofilms living on gold grain surfaces (Craw and Lilly, 2016; Fairbrother et al., 2012, 2013; Freise, 1931; Reith et al., 2010, 2006). Laboratory studies have demonstrated that bacteria, when exposed to mobile gold complexes, will form nanophase gold particles such as colloids and euhedral crystals (Fairbrother et al., 2013; Reith et al., 2009). During biofilm growth and development, organic materials can trap and aggregate these nanoparticles and form millimeter-scale grains (Shuster and Southam, 2014; Southam and Beveridge,

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1994). From the surface of natural gold grains, concavities have been interpreted as 'corrosion pits' formed by the dissolution of gold and silver (Colin et al., 1989; Larizzatti et al., 2008). Recent studies by Craw and Lilly (2016) highlighted that thiosulfate complexes may have an important role in gold mobility that can lead to subsequent nugget formation. Furthermore, gold mobility has been attributed to seasonal evaporation and changes to groundwater levels in hot arid environments (Hough et al., 2008). In combination, these previous studies highlight the dynamic conditions of secondary gold grain transformation in supergene environments. However, environmental rates of these biogeochemical transformation processes are unknown. Therefore, the purpose of this

study is to estimate the kinetics of gold biogeochemical cycling with respect to the formation of colloidal gold and their mobility within a placer environment. To achieve this, results from structural and chemical analyses of gold colloids occurring on the surface of gold grains were used to calculate temporal estimates of gold dissolution/re-precipitation. In addition, experiments involving the adsorption of colloidal gold onto fluvial sediment were performed to assess the dispersion of nanophasic particles within a hydrodynamic environment. This study highlights the important role of materials characterization of natural, nanophasic gold particles for interpreting biogeochemical processes of gold mobility and transformation in subtropical environments.

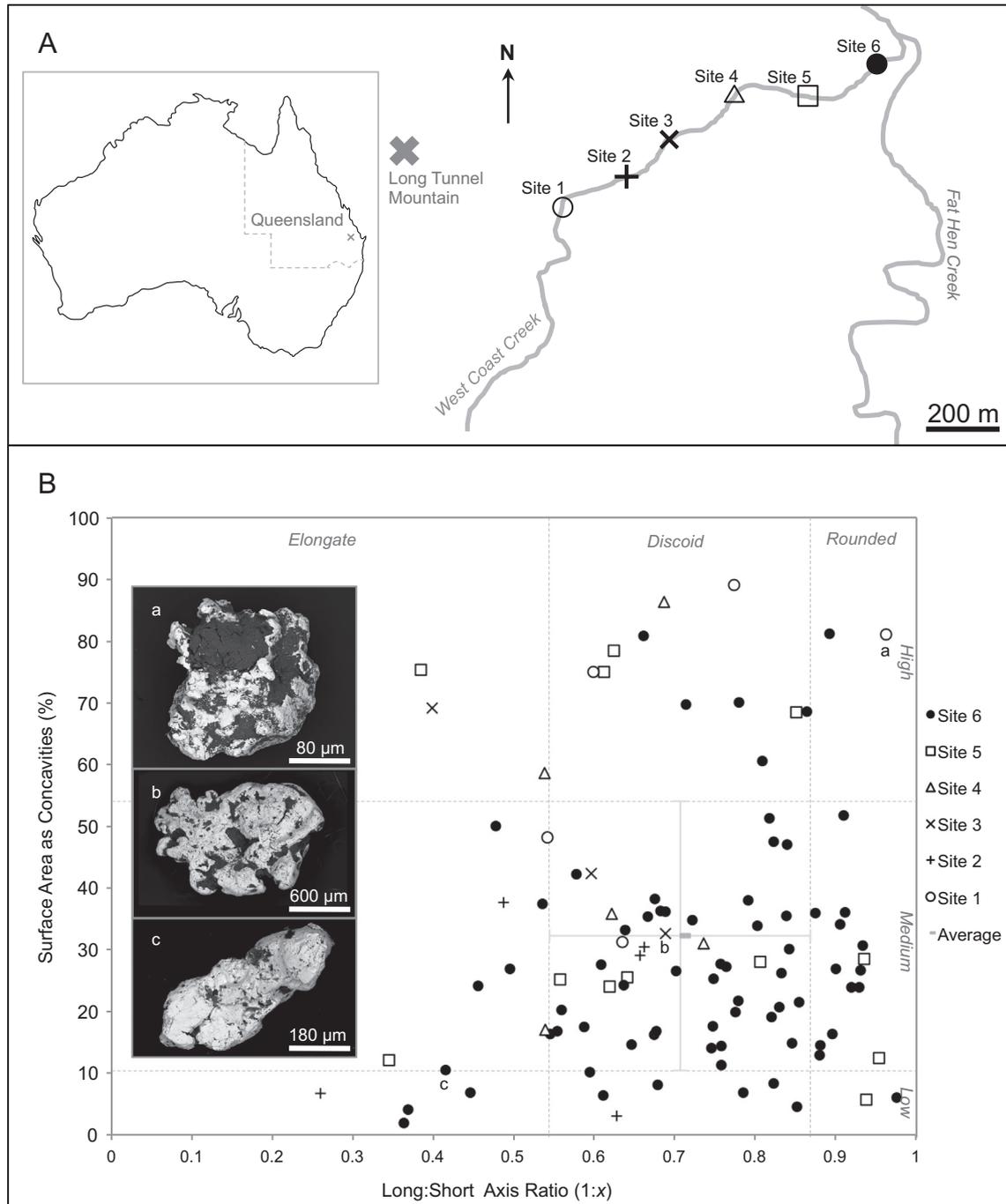


Fig. 1. A map of sampling locations at West Coast Creek, Queensland, Australia (A). A plot of long to short axis ratio of each grain and the percentage of surface area occurring as concavities filled with sediment. Dashed lines represent standard deviations of the long to short axis ratio and the concavity surface area. As a group, gold grains demonstrated variations in morphology and the extent of weathered surfaces (B).

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