Biogeochemical cycling of gold: Nanoparticle formation and accretion

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The biosphere catalyzes a variety of biogeochemical redox reactions resulting in the transformation of gold (Au). In particular, the oxidation and reduction of sulfur compounds appears to drive the cycling of Au in many environments. Weathering of electrum produces 'bacteriomorphic' Au structures, a controversial form of 'biogenic' Au representing a dissolution feature of hypogene material. From the geochemistry literature, weathering of Au-bearing metal sulfides produces chemically mobile Au(I)-thiosulfate-, and in hypersaline environments, Au(III)-hydroxo-chloride complexes. Conversely, microbial destabilization of these Au(I/III)-complexes is coupled to the biomineralization of Au, *i.e.*, the formation of secondary Au, completing the cycle, and potentially forming genuine bacteriomorphic gold. However, the biogeochemical transformation of Au does not stop here. Previously, Friese [1] highlighted the importance of biodegradation of plant materials, presumably mediated by microbial activity, for the formation of macroscopic aggregates of crystalline secondary Au. Using culture studies, we have demonstrated that destabilization of Au(I/III)-complexes can occur directly and indirectly by the growth of bacteria and through microbial Fe- and S-cycling. Synchrotron spectroscopy techniques (μXRF, μXANES and μEXAFS) have allowed us to map the distribution and speciation of Au in individual *C. metallidurans* cells and demonstrate the importance of organic sulfur compounds in the transformation of Au(I/III). In these systems, secondary Au begins as colloidal nano-particles that aggregate forming micro-crystalline octahedral Au crystals. Studying these interactions from the nano- to the macro-scale has shown a close correlation between laboratory culture experiments and field materials, where the continuum of dissolution – re-precipitation processes can result in the formation of laboratory or naturally 'grown' Au grains. These grains represent an increase in size between 5 and 6 orders of magnitude compared to the 'original' colloidal material. Remarkably, through microbial action, aggregation of nano-phase gold into gold grains can occur on the time-scale of days.

[1] Freise. 1931. *Economic Geology* ***26***, 421-431.