7. Conclusions and Implications

One of the major contributions of this thesis is our finding that phylogenetically distinct Fe(II)-oxidizing phototrophs fractionate Fe isotopes via apparently equilibrium processes. While our work could not distinguish between an equilibrium fractionation mediated by biological or abiotic processes, it demonstrated that equilibrium processes prevail in biological systems rather than kinetic process as previously hypothesized [10]. Further the likely possibilities we provided for the mechanism of Fe isotope fractionation by these organisms guided subsequent investigations where our hypothesis that the fractionation represented equilibrium exchange between aqueous Fe(II) and Fe(III) species overlain by kinetic effects produced by precipitation of ferric minerals was proven true [8]. Finally, while we found that these organisms fractionate Fe isotopes in a way that is consistent with Fe isotopic values found in Precambrian BIFs, we concluded that it is unlikely that this fractionation can be used as a biosignature for this metabolism given its similarity to fractionations produced by abiotic Fe(II) oxidation reactions, thus, making further study in this area largely unnecessary.

Organisms that oxidize Fe(II) are difficult to study from a genetic perspective. This is largely due to the challenges inherent in growing these organisms. For example, aerobic neutrophilic Fe(II)-oxidizers must outcompete the rate of abiotic oxidation of Fe(II) by molecular oxygen (O_2) to harvest energy for growth. The requirement of specific O_2 and Fe(II) concentrations for these

bacteria is met by growing them in tubes of solid medium with opposing gradients of Fe(II) and O_2 [54]. Such culturing requirements are not easily amenable to large scale genetic screens. Thus, a second contribution of this work is the development of an assay for the identification of genes involved in Fe(II) photoautotrophy in genetically intractable strains. With this assay, we are afforded a means to identify the molecular components of Fe(II) oxidation and using this assay we have identified the first genes known to be involved in this metabolism. Future work to identify additional components of this metabolism are now enabled and should include the identification of the enzyme that catalyzes Fe(II) oxidation, its localization in the cell, and investigations of the degree to which this enzyme is conserved among phylogenetically distinct organisms able to oxidize Fe(II). We anticipate that such phylogenetic investigations will provide insight, not only into the mechanism of this metabolism, but also its origins.

Overall, this thesis provides an example of what one might call “metabolic paleontology,” that is: the investigation of the mechanisms of modern metabolisms as a means to uncover how ancient related metabolisms may have affected the geochemical evolution of the Earth. It is important to note, however, that a fundamental assumption in this work is that the metabolisms of modern microbes are representative of those of ancient organisms. The uncertainty in this assumption is irresolvable, as we can never know to what extent ancient metabolisms may differ from modern metabolisms. However, a thorough understanding of the molecular components of a particular metabolism of interest

and how their expression is regulated in a diversity of modern organisms can help to reduce this uncertainty by giving us a feel for the range of variability in the rates and particular components of these metabolisms. Moreover, with molecular investigations, we can uncover the degree to which aspects of this these metabolisms have been conserved throughout their evolution, as it is in these aspects were our most robust conclusions can be drawn. With such comprehensive comparative studies we can make progress towards the identification and unambiguous interpretation of biosignatures in the rock record.

Given the extensive time-period over which BIFs were deposited, it is probable that a combination of biotic (both an- and oxygenic) and abiotic mechanisms contributed to the deposition of these rocks and that the relative contributions of biotic and abiotic Fe oxidation varied over geologic time. Therefore, it is important to note that a model for the contribution of Fe(II) oxidizing phototrophs in the deposition of BIFs does not preclude a role for abiotic mechanisms of Fe(II) oxidation and recognize that an understanding of the chemistry of the Earth at the time of BIF deposition is critical in determining which Fe(II) oxidizing metabolisms were involved in the formation of BIFs over time. Furthermore, such temporal variations in the role of direct biological and inorganic processes may produce identifiable morphological or chemical variances in BIFs of different ages and may present a potential target for biosignature development.

In conclusion, studying extant microbes to identify chemical signatures unique to these organisms may provide us with tools to investigate how the

metabolism of ancient related organisms shaped the chemistry of the Earth. Although I was not able to identify a biosignature unique to the metabolism of these organisms, my research makes significant contributions toward this lofty goal and it is my hope that these investigations will lay the groundwork for future studies with this directive.