Changes in carbon oxidation state of metagenomes along geochemical redox gradients

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Elucidating the linkages between biomolecules and environments is a major goal for geobiochemical studies of natural microbial communities. As shown by next-generation metagenomic and metatranscriptomic sequencing, geochemical redox gradients strongly impact the structure and likely metabolic activity of communities. However, the information contained in these datasets about the chemical composition of biomolecules has not been explicitly studied. We calculated the average oxidation state of carbon (Z_C) to test the hypothesis that geochemical redox gradients have a systematic influence on the chemical compositions of DNA, RNA, and proteins. A survey of Z_C was conducted for published metagenomic and metatranscriptomic datasets representing geochemical redox gradients associated with depth transects in oxygen minimum zones (OMZ) of oceans, other aquatic environments including hypersaline lakes, cooling and mixing of terrestrial or submarine hydrothermal fluids, near-surface phototrophic and deeper layers of a microbial mat, a hydrocarbon-rich mud volcano, and sediment subsurface environments. The Z_C of DNA and putative proteins parallels measured oxygen concentrations for mixing between hydrothermal and oxygenated fluids and in near-surface zones of oceans and the mat, and also increases with proximity to the surface in the mud volcano and in one anoxic sediment dataset. In contrast, Z_C increases with depth in OMZs and the deep layers of the mat despite the low oxygen concentrations in these settings. These trends may reflect distinct processes responsible for the development of the gradients, such as addition of O_2 by mixing of fluids, or depletion of O_2 and other electron acceptors by internal processes including microbial respiration. Although Z_C of DNA is generally positively correlated with that of putative proteins in individual datasets, a separation of different global environments is more evident on a cross-plot of data for proteins and DNA. These results reveal systematic relations between the carbon oxidation state of biomolecules and geochemical redox gradients at both local and global scales, which can inform more strongly integrated thermodynamic and phylogenetic models of the evolution and geobiochemistry of microbial communities.