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Intermediate sulfur products along natural marine redox gradients: new insights from a holistic analysis of microelectrode cyclic voltammograms

MUSTAFA YÜCEL[1]*

[1] Middle East Technical University, Institute of Marine Sciences, METU Erdemli Campus, Mersin, Turkey; (*correspondence: muyucel@metu.edu.tr)

Seafloor dissolved sulfide gradients are prevalent features in coastal and productive sea basins. In deep anoxic basins they may occur over 100s of meters whereas in sediments and microbial mats the scales of these gradients shrink down to millimeters. Here I will comparatively analyze a large set of voltammetric scans from sulfide gradients from the Black Sea, Baltic Sea and benthic microbial mats for insights into their potential to generate sulfur intermediate products such as electrochemically (gold-amalgam voltammetry) detectable polysulfide and iron monosulfide species. In the Baltic Sea, a mostly hypoxic basin, the presence of these intermediates occurs in spatially distinct zones along a sedimentary redox gradient, pointing to a polysulfide-yielding oxidation pathway within benthic microbial mats but a FeS-yielding - probably abiotic process in deeper sediments. This explicit detection of FeS is well corroborated by the shifting reverse to forward cyclic voltammetric signal ratio related to sulfide deeper in the core. In a similar benthic redox gradient in the Black Sea, an ongoing sulfidation-related pathway in deeper part of the upper sediment column is only implicit in the changing ratios of magnitudes of sulfide-dependent signals during forward and reverse scans. The lack of a distinct FeS signal here either points to larger than 100 micrometer (hence not electroactive) dissolved nanoparticles and/or organic sulfur species. As a result, microelectrode cyclic voltammetry is not only useful for detecting individual analytes but a comparative analyses of reverse and forward scans can yield more information on processes involving sulfur intermediates.