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Authigenic pyrite mediated by extremophile microorganisms related with active methane-seeps in the Gulf of Cadiz: evidences from textural, geochemical and underwater observations

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Methane-seeps in the Gulf of Cadiz are frequently accompanied by carbonate and sulphide precipitates. Sulphide crusts and aggregates, and sediments were sampled by coring and benthic dredges during the Anastasya-01 and TTR-15 cruises at flanks and tops of extrusive structures (mud volcanoes and diapiric ridges), located on the shelf and slope of the Gulf of Cadiz. In the northern sector we recovered pyrite crusts in the Guadalquivir Diapiric Ridge (GDR), and pyrite-marcasite crusts and aggregates were sampled in the Moroccan Margin (MM). Underwater photograph tracks and videos were also acquired across the sampled areas using a *Benthos-372* camera and a TV-

grab respectively. Video observations and photographs show textural, biological and colour differences between the areas affected by seeps (active or inactive actually) and the surrounding sediments, not affected by fluid venting. Active seep zones are characterised by bubbling and superficial sediment olive-white in colour covering surfaces of variable size, where are present chemosynthetic fauna as *Phogonophora sp*. *Calyptogena*, *sp*., and *Acharax sp*. The filamentous textures visible in sediments from these areas suggest presence of sulphide oxidising bacterial mats (*Beggiatoa sp*., *Arcobacter sp*.?). Inactive seep zones (without brine flowing across) are composed by sediments with intense orange-red colour and abundance of Fe-oxides aggregates, probably mediated by *Leptothrix ochracea* and *Beggiatoa sp*. These two types of bacterial mats were in close proximity.

Sulphide crusts recovered around the gas emission sites on the sea floor in the GDR are basically composed by pyrite cementing detrital grains of sediments. Pyrite appears in two habits, as framboids and framboidal aggregates (formed by multiple octahedral or pentagon-dodecahedral pyrite micro crystals) or massive, generally surrounding framboidal pyrite. Organic matter around or within pyrite micro crystals was observed. Sulphur isotopic ratio ($\delta\ ^{34}S$) measured for one pyrite crust from this area was –41per mil CDT. Pyrite aggregates sampled within the sediment column in the MM are formed by cubic idiomorphic crystals from millimetres to centimetres. They contain residual grains of silicates and the matrix of these samples is mud breccia (olive-grey in colour) with presence of H_2S. Sulphur isotopic ratios for the sulphides from the Moroccan sector ($\delta\ ^{34}S$) ranging from + 25.3 to + 4.3 per mil CDT. All the samples recovered in both sectors GDR and MM present lack of oxidation and therefore goethite or other Fe-oxides are practically absent. Sulphides are enriched in trace metals as Mo, Pb, Ni, and Co.

The sediments are affected by seepage of hydrocarbon-rich fluids, which induced intergranular precipitation of authigenic pyrite and carbonates mediated by chemosynthetic microbial communities, which consist of methanotrophic archaea and sulphate-reducing bacteria (SRB) mediating anaerobic oxidation of methane. Sulphides studied in this work represent biologically induced biomineralizations, where pyrite and marcasite were precipitated by indirect effect of SRB methabolism around focused or diffuse methane-seeps. At GDR the pyrite crusts precipitation could have taken place by successive processes of bio oxidation-reduction by sulphur-disproportionating bacteria. Repeated cycling of sulphide oxidation followed by sulphur disproportionation, could lead to high sulphur isotope fractionations observed here. Samples from the MM were formed within the sediment column, in a confined ambient whit limited supply of sulphate from the sediment-water interface.