

## **Sulfur and iron stable isotope and trace element patterns of pyrite record sulfate-driven anaerobic oxidation of methane**

ZHIYONG LIN<sup>1,2\*</sup>, XIAOMING SUN<sup>1</sup>, HARALD STRAUSS<sup>2</sup>,  
REINER KLEMD<sup>3</sup>, KAIYUN CHEN<sup>4</sup>, JÖRN PECKMANN<sup>5</sup>

<sup>1</sup> School of Marine Sciences, Sun Yat-sen University,  
Guangzhou, China (\*linzhiy9@mail.sysu.edu.cn)

<sup>2</sup> Institut für Geologie und Paläontologie, Westfälische  
Wilhelms-Universität Münster, Münster, Germany

<sup>3</sup> GeoZentrum Nordbayern, Universität Erlangen,  
Schlossgarten 5a, 91054 Erlangen, Germany

<sup>4</sup> State Key Laboratory of Continental Dynamics, Department  
of Geology, Northwest University, Collaborative  
Innovation Center of Continental Tectonics, Xi'an  
710069, China

<sup>5</sup> Institut für Geologie, Centrum für Erdsystemforschung und  
Nachhaltigkeit, Universität Hamburg, Hamburg, Germany

In areas of methane seepage<sup>[1]</sup>, most of the methane is oxidized in the sediment at the expense of sulfate by a biogeochemical process known as the sulfate-driven anaerobic oxidation of methane (SO<sub>4</sub>-AOM)<sup>[2]</sup>. During this process, hydrogen sulfide is released and eventually results in the formation of pyrite when sufficient iron is available<sup>[3,4]</sup>. Herein, we present the sulfur, iron isotopic and trace element compositions of authigenic pyrite from seafloor sites in two seepage areas of the South China Sea. Both sulfur and iron isotopic compositions of pyrite reveal similar patterns with depth in both areas, displaying an extreme variability in  $\delta^{34}\text{S}$  values ranging from  $-50$  to  $+110\%$ , and a large range in  $\delta^{56}\text{Fe}$  values falling between  $-1.0$  and  $+1.5\%$ . Interestingly, the enrichment of  $^{56}\text{Fe}$  in pyrite at certain depths coincides with extremely high  $\delta^{34}\text{S}$  values of pyrite. In addition, some trace elements (e.g., Mn, Pb, Zn) in these pyrites reveal abnormal enrichments. These observations indicate that pyrite formation resulted from SO<sub>4</sub>-AOM at the sulfate-methane transition zone. During SO<sub>4</sub>-AOM, the enhanced production of hydrogen sulfide promotes reductive dissolution of iron-manganese (oxyhydr)oxides in the sediment, accelerating the release of trace elements into the pore water, which are subsequently incorporated into pyrite. Therefore, the combined enrichments of  $^{34}\text{S}$  and  $^{56}\text{Fe}$  and some trace elements in pyrite may represent a tracer for SO<sub>4</sub>-AOM in modern and ancient sedimentary sequences.

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