

***In situ* detection of submarine hydrogen using deep-sea Raman spectroscopy**

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Hydrogen, as a clean and sustainable energy source, has garnered significant attention for its potential to mitigate climate change and contribute to the global transition towards renewable energy. Recent studies have highlighted the significance of natural hydrogen sources, especially in submarine environments where serpentinization and microbial activity contribute to hydrogen generation. This study presents an innovative approach to the *in situ* detection of submarine hydrogen using deep-sea Raman spectroscopy, focusing on two distinct hydrothermal systems: the east Caroline plate in the western Pacific and the Qianyuan vents in the Okinawa Trough.

In the east Caroline plate, a large-scale hydrothermal field was identified with hydrogen-rich fluids supporting a diverse chemolithoautotrophic ecosystem. Raman spectroscopy on-site revealed hydrogen concentrations of 5.9–6.8 millimoles/kilogram, offering direct evidence of hydrogen release from serpentinization reactions in deep-sea hydrothermal systems. The total hydrogen flux from this site was estimated to exceed 5% of the global submarine abiotic hydrogen flux, suggesting that current estimates may be significantly underestimated.

Similarly, the Qianyuan vents in the Okinawa Trough, characterized by high hydrogen fluxes of approximately 1,000 tonnes per year, were investigated using *in situ* Raman analysis. The gas composition of the hydrogen-rich fluid mixture—comprising 70.2% CO₂, 17.3% CH₄, 10.6% H₂, and 1.9% H₂S—demonstrated the potential for microbial and serpentinization-driven hydrogen production. The study also identified the presence of hydrogen-rich gas hydrates, supported by seismic data indicating significant hydrogen accumulation beneath the seafloor.

The application of Raman spectroscopy in these extreme environments allows for the precise characterization of hydrogen concentrations and gas compositions without the need for sample collection, providing invaluable data for understanding natural hydrogen reservoirs in marine settings. This research demonstrates the potential of back-arc basins and deep-sea hydrothermal fields as critical sites for natural hydrogen production and accumulation. The findings suggest that deep-sea environments, particularly those influenced by serpentinization, could offer new opportunities for exploring hydrogen as a viable marine energy resource.

