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In situ Raman-based detections of the hydrothermal vent and cold seep fluids

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Hydrothermal vents and cold seeps, and their associated biological communities play an important role in global carbon and sulphur biogeochemical cycles. Most of the studies of fluid composition geochemistry are based on recovered samples, both with gas-tight samplers and as open specimens, but the *in situ* conditions are difficult to maintain in recovered samples. Determination *in situ* of the chemical signals of the emerging fluids are challenging due to the high pressure, often strongly acidic and temperature in which few sensors can survive. Most of those sensors used so far are based on electrochemistry, and can typically detect only a few chemical species.

Here we show that direct measurement of critical chemical species of hydrothermal vents and cold seeps can be made rapidly and *in situ* by means of a new hybrid version of earlier deep-sea pore water Raman probe carried on the ROV (Remote Operated Vehicle) *Faxian*. The fluid was drawn through the probe by actuating a hydraulic pump on the ROV, and measured at the probe optical cell through a sapphire window.

We have observed the concentrations of H_2S, HS^-, SO_4^{2-}, HSO_4^-, CO_2, and H_2 in hydrothermal vent fluids from the Pacmanus and Desmos vent systems in the Manus back-arc basin, Papua New Guinea. Two black smokers (279˚C and 186˚C) at the Pacmanus site showed the characteristic loss of SO_4^{2-}, and the increase of CO_2 and well resolved H_2S and HS^- peaks. At the white smoker of Onsen site the strong HSO_4^-peak observed at high temperature quickly dropped with strong accompanying increase of $SO_4^{2-}$and H_2 peaks when the sample contained in the Raman sensing cell was removed from the hot fluid due to rapid thermal deprotonation. We report here also the finding of a new lower temperature (88˚C) white smoker “Kexue” field at the Desmos site with strong H_2S, HS^- and CO_2 signals.

We also have detected the concentrations of CH_4,H_2S, HS^-, SO_4^{2-}, and S_8 in cold seep fluids and the surrounding sediment pore water from the northern South China Sea. Several sediment pore water profiles nearly at the cold seep vent showed the characteristic loss of SO_4^{2-}, and the increase of CH_4, H_2S and HS^- peaks. Dissolved S_8 and CH_4had been first found at the fluids under the lush biological communities of the cold seep. This may indicate some bacteria mats at the lush biological communities oxidize hydrogen sulfide and produce elemental sulfur as a byproduct.

Our research suggests that the *in situ* observed H_2S:HS^-, and HSO_4^-:SO_4^{2-} ratios provide elegant pH sensitive “dyes” with which to diagnose the geochemical reactions occurring.