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Can clumped isotopes help us constrain the origin of H_2?

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Dihydrogen (H_2) is becoming a key molecule for the energy transition. Many processes can lead to the formation of H_2 such as methane reforming, water electrolysis in the industry or hydrothermal alteration, radiolysis, volcanism and microbial processes in nature. Tracing the origin of H_2 in natural settings remains challenging. Bulk hydrogen isotopes provide some clues to distinguish high temperature H_2 from low temperature H_2, or to constrain whether a system is at the equilibrium or not. However, this signature does not lead to distinguish one type of H_2 from another.

H_2 clumped isotopes may allow constraining the origin of dihydrogen. This approach was pioneered by Popa et al. [1] on a MAT253-ULTRA. Natural gas samples were measured by Mangenot et al. [2], revealing that H_2 clumped isotopes appear re-ordered at environmental temperatures in most settings. However, all natural samples studied so far were hydrothermal gases, where hydrogen bonds had the potential to be activated by metal catalysts and re-ordered at environmental temperatures. Under those circumstances, H_2 clumping would not keep a record of dihydrogen synthesis.

Here we investigate on a collection of natural and experimental samples the potential for H_2 clumped isotopes to have escaped re-ordering to environmental temperatures. After a review, about the use of bulk hydrogen isotopes and their implications, we will present the process of clumped isotopes measurement, beginning with the purification of H_2 from other gases that can affect the fractionation of H_2. Based on the available data, we will finally present the preliminary results on the measurements of clumped isotopes of H_2 on the MAT253-ULTRA installed at IPGP.

[1] Popa, Paul, Janssen, and Röckmann (2019), *Rapid Communications in Mass Spectrometry* 33, 239–251.

[2] Mangenot, Xie, Crémière, Giunta, Lilley, Sissmann, et al. (2023), *Chemical Geology* 621, 121278.