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Pushing paleoclimate and petroleum research forward: A perspective on the various applications of HR-IRMS

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Classical stable gas isotopes, such as δD, $\delta^{13}C$, $\delta^{18}O$ are well-established and valuable tools in various fields of earth sciences. They aid in constraining genetic mechanisms, provenance, transport processes, and formation temperatures. Their geothermometric usability is however limited as they lack an absolute temperature dependency. Clumped isotope geochemistry overcomes this limitation since the equilibrium distribution ('clumping') of heavy isotopes within multiply substituted molecules is solely dependent on the formation temperature [1].

The most evolved clumped isotope applications are clumping in CO_2 and in CH_4 and it has been widely demonstrated that associated geothermometry can be used to reliably determine highly precise carbonate and methane formation temperatures. When two clumped isotopologues of one species are analyzed, it is even possible to pin down kinetic biases in cases where a sample is deviating from the expected thermodynamic equilibrium.

Apart from geothermometry and the characterization of non-equilibrium processes, clumped isotopes offer additional constraints on forensic source apportionment of natural gas samples. Recently, the classical isotope toolbox has been expanded by novel clumped isotope signatures (in methane: $\delta^{13}CH_3D$ and $\delta^{12}CH_2D_2$ [2]; in carbon dioxide: δ_{47} and δ_{48} [3]; in nitrogen: δ_{30} [4]; in hydrogen: δDD [5]).

In this presentation I will give examples to emphasize how clumped isotopes in a variety of gases (e.g. CO_2, CH_4, N_2, H_2) add irreplaceable value and fresh insights into petroleum and paleoclimate research. I will also highlight the benefit of direct analysis of triple oxygen isotopes in carbonate CO_2 by HR-IRMS [6] to clumped CO_2 measurements.

[1] Eiler (2007) *EPSL 262*, 309-327 [2] Eldridge et al. (2019) ACS Space Chem, doi:10.1021/9b00244 [3] Bajnai et al. (2020) *Nat Commun 11*:4005 [4] Yeung et al. (2017) *Sci Adv 3*:eaao6741 [5] Popa et al. (2018) *RCMS*, doi: 10.1002/rcm.8323 [6] Adnew et al. (2019) *RCMS*, doi: 10.1002/rcm.8478