## Origin of H<sub>2</sub> and CH<sub>4</sub> gases in the Eastern São Francisco Basin, Brazil

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This multi-year study reveals a series of systems rich in He and H<sub>2</sub> gas, and a complex CH<sub>4</sub> cycle with multiple abiotic and biological sources. Surface gas seeps along rivers are dominated by microbial CH<sub>4</sub> related to near-surface processes in tropical sediments and wetlands. In contrast, samples from gas exploration wellheads have He (up to 1% by vol) and high concentrations of H<sub>2</sub> (up to 25-40% by vol. but consistently between at least 7-11%) for samples measured between 2012 and 2019. Here for the first time an exploration gas well discharging at surface shows evidence of the type of deep cratonic gases typically associated with the deep mines of the Witwatersrand Basin and Canadian Shield<sup>1</sup>. Specifically, some wells show a significant component of abiotic alkanes with high associated concentrations of H<sub>2</sub>, higher hydrocarbons, and isotopic and geochemical characteristics associated with abiotic organic synthesis<sup>2</sup>. Noble gas analyses confirm a crustal rather than mantle source. Particularly notable is the elevated <sup>21</sup>Ne/<sup>22</sup>Ne endmember identified in at least one gas well, and from other areas of the São Francisco Craton<sup>3</sup>, that both show the characteristic elevated neon end-member value first identified in ancient fracture fluids from deep mines in Canada and South Africa<sup>4,5</sup>.

Overall, these results demonstrate the H<sub>2</sub>-rich gases in the Precambrian to early Paleozoic cratonic rocks of Brazil share important characteristics with the deep gas and ancient fluids first described in the deep mines of the Canadian Shield and Witwatersrand Basin. The exploration gas wells reflect a complex mixture of discharging gas associated with abiotic organic synthesis and H<sub>2</sub> production (likely related to radiolysis and/or serpentinization), and local mixing with what are likely more surficial sources of microbial CH<sub>4</sub>.

<sup>1</sup>Sherwood Lollar et al. (2021) GCA 294:295-314. <sup>2</sup>Warr et al. (2021) GCA 294:315-334. <sup>3</sup>Magalhães et al, Goldschmidt Abstract (2021). <sup>4</sup>Lippmann-Pipke et al. (2011) Chem Geo 283:287-296. <sup>5</sup>Holland et al. (2013) Nature 497:357-360.