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Thermodynamic and kinetic constraints on molecular hydrogen abundance and isotope systematics in hydrothermal fluids

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Molecular hydrogen is one of the major gas constituents in hydrothermal fluids where it plays a pivotal role in geological and biological processes. Nonetheless, factors controlling molecular hydrogen abundance and isotope fractionation remain poorly understood and quantified. We report hydrogen fugacity (fH_2) and δD of H_2 and H_2O for hydrothermal fluids of variable temperature (226-359 °C) from terrestrial volcanic arc and rift systems sourced by seawater and meteoric water The hydrothermal fluid fH_2, δD-H_2 and δD-H_2O values are of 0.002-3.3 bar, -646 to -391‰ and -94.1 to +11.3‰, respectively. Comparison of dataset with results of geochemical modeling revealed that in meteoric water systems H_2 production is controlled by the reduction of H_2O upon oxidation of aqueous Fe^{II} to Fe^{III} and subsequent formation of Fe-containing mineral. Elevated sulfur contents, sourced from volcanic gas and/or seawater, result in oxidation of aqueous S^{-II} to pyrite and S^{+VI}, providing a further source of electrons along with Fe^{II} oxidation. Our results show that hydrogen fugacity in hydrothermal fluids is controlled by metastable equilibria along a fluid-rock reaction path that primarily depend on temperature, rock-to-water ratio, source water composition and volcanic gas input. The dataset demonstrates that δD ratio of H_2 is controlled by the isotopic composition of the source water and equilibrium isotope fractionation at the hydrothermal reservoir temperatures. Upon fluid ascent to surface, H_2-H_2O isotope exchange re-equilibration may occur, this depends on cooling rate coupled with the kinetics of the isotope fractionation reaction. Based on the extent of hydrogen isotope disequilibrium, we estimate reservoir-to-surface travel times of minutes to <3 hours and few hours to days for geothermal wells and fumaroles fluids, respectively. The Project has received funding from the European Union’s Horizon 2020 under Grant Agreement #818169.