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Pressure-Induced Variations and Reversal of Hydrogen Isotope Fractionation During Slab Dehydration

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Subduction represents the primary pathway by which surface water is transported into Earth's interior. During this process, hydrous minerals undergo dehydration and phase transformations, inducing significant hydrogen isotope fractionation. Quantifying H isotope fractionation under high-pressure and high-temperature conditions is therefore critical for constraining the hydrogen isotopic composition of water entering the deep mantle. However, experimental constraints on H isotope fractionation among hydrous minerals and aqueous fluids at elevated pressures and temperatures remain limited. Here, we combine deep potential models with path-integral molecular dynamics (PIMD) simulations to investigate hydrogen isotope fractionation between water and major subduction-zone hydrous minerals over a wide P–T range (300–1273 K, 0–8 GPa). The studied phases include serpentine, chlorite, glaucophane, phengite, lawsonite, and epidote. Reduced partition function ratios (RPFRs) and equilibrium isotope fractionation factors were calculated to evaluate mineral–fluid hydrogen isotope fractionations under subduction conditions. Our results reveal contrasting pressure dependencies among different hydrous phases. For most hydrous minerals, (e.g., serpentine and chlorite), the ability to enrich deuterium (as reflected by increasing RPFR values) strengthens with increasing pressure. In contrast, water, epidote, and lawsonite exhibit decreasing RPFR values with pressure, indicating reduced D enrichment at higher pressures. These opposite trends are primarily controlled by variations in hydrogen-bond length and strength within each structure, which respond differently to compression. Moreover, hydrogen isotope fractionation between minerals and coexisting fluids shows a pronounced pressure effect: with increasing pressure, mineral–fluid fractionation factors generally increase and may even reverse at high pressures. These findings suggest that dehydration reactions during slab subduction do not necessarily remove isotopically heavy hydrogen from the slab. Instead, fluids transported into the deep mantle may exhibit relatively high δD values. This study provides the first systematic constraints on hydrogen isotope fractionation in hydrous minerals across subduction-zone P–T conditions and offers new insights into interpreting hydrogen isotope signatures in Earth's deep water cycle.