

Active silicate alterations and its role in major cation sinks in volcanic ash-rich Hikurangi Margin sediments

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This study used porewater and sediment samples of Site U1518 drilled on the Hikurangi Margin during IODP Exp. 375 to investigate sedimentary silicate dynamics associated with volcanic ash alteration and its interactions with alkalinity and major cations. We applied a sequential chemical leaching approach in combination with stable Si isotope and mineral analysis. We observed that porewater $d^{30}\text{Si}$ values decrease from +0.63 to -0.5 ‰ with increased dissolved silica (DSi) concentrations of up to 751 mM in the upper 20 mbsf, suggesting active volcanic ash dissolution. This is so far the lowest reported porewater $d^{30}\text{Si}$ value. From 20 down to 160 mbsf, porewater $d^{30}\text{Si}$ values gradually increase to +1‰ accompanied by a slightly increased DSi concentrations from 541 to 647 mM. Preliminary results of sediment leaching show the $d^{30}\text{Si}$ values of HCl leachates ranging from -3 to -1 ‰. Meanwhile, the HCl leachate of volcanic layer samples showed gradually decreased $d^{30}\text{Si}$ values along with continuous Al, Fe, Mg dissolving into supernatant within a 24 h leaching period, possibly due to solubility of different cation-rich silicate phases in acid. This also suggests that the majority of Mg and Fe sinks in such sediments occurs at the precursory stage of authigenic clay formation. The $d^{30}\text{Si}$ values of Na_2CO_3 leachates vary from +0.09 to +1.1 ‰ in the volcanic ash layer. The variation could be primarily attributed to neoformed clay minerals, which preferentially incorporates lighter Si isotopes, and to a lesser extent biogenic silica (BSi) dissolution given the low downcore BSi contents between 0.14 – 0.3% and low Si/Al ratios of 5 - 10 in the Na_2CO_3 leachates. The $d^{30}\text{Si}$ values of the NaOH leachates in the volcanic ash layer are around -1– -0.5 ‰, which fall into the range of those reported values for aged clay minerals but slightly lower than primary silicates. A reactive-transport model will be developed to simulate reaction rates of different silicate phases. Our results show that volcanic ash alteration can result in complex Si re-distribution among a variety of silica and silicate minerals, directly linking to alkalinity and major ion budgets in the Hikurangi Margin sediments.