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Linking reactive silicate phases and porewater signatures in deep sea sediments from Western Antarctica

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In the present work, we investigate marine porewater and sediment leachate geochemistry from an 800m long sediment core in the Amundsen Sea, dating back to Miocene (5.7 Ma). The sediment lithology is dominated by silty clay, containing dispersed biogenic silica (bSi) and the depositional environment receives a high influx of terrigenous sediments through glacial supply. A bulk XRD analysis reveals dominance of illite-smectite-mica type clays (28-42%), followed by quartz and plagioclase. The dissolved silicon profile of porewater (DSi_{pw}) varies between 60 and 900 mM, with three distinct zones of high DSi_{pw}. These zones are associated with abundant biosiliceous deposits (diatoms and radiolarians), high Si/Al ratio in sediments and high bSi (wt%). Thus, zones of high DSi_{pw} points to a pelagic and/or hemipelagic sedimentation, associated with ice sheet retreat during interglacial periods. In order to distinguish sources of DSi_{pw}, we employed a sequential leaching technique to separate reactive Si phases from sediments.

The bSi leached during the first hour of Na_2CO_3 exhibits a higher Si/Al ratio and $d^{30}Si$ values ranging from 0.6 to 1.5‰, consistent with diatoms and radiolarians signatures in the Southern Ocean. We observe a positive correlation between $\delta^{30}Si$ values and Si/Al and Si/K ratios within the Na_2CO_3-1hr leachates, with higher Si/Al ratio associated with high bSi (wt%) and DSi_{pw} and vice versa. Thus, 'altered bSi' represented in Na_2CO_3-1hr leachates with low $\delta^{30}Si$ values, and high Al and cation content, can be sourced from concurrent leaching of amorphous aluminosilicates and/or detrital silicates in the sediments. The HCl leachate fractions represent the lightest Si pool, with $d^{30}Si$ varying between -3.1 to -0.7 ‰, sourced from amorphous authigenic aluminosilicates and/or metal oxide. Variability within the HCl leachates is influenced by the Mg, K and Fe contents, with more depleted ^{30}Si associated with higher Mg/Al, K/Al and Fe/Al ratios. In contrast, the strong alkaline leaching with 4M NaOH represent the largest Si pool (188-830 mmolSi g^{-1} sediment) with a homogenous $d^{30}Si$ value (-0.1±0.3‰), consistent with a dominant lithogenic Si source. Overall, the marine silicate alterations in sediments are intrinsically linked to supply of reactive silicates, controlled existing environmental conditions.