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Extremely high alkalinity due to dissolution of mica-group silicate in the pelagic sediments of the Ulleung Basin (East Sea): stable Si isotopes evidence and reactive transport modelling

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Marine silicate alteration is a combined process of lithogenic silicate (LSi) dissolution (known as marine silicate weathering) and secondary clay neoformation (known as reverse weathering). Both processes have been shown to affect long-term C cycling. Yet, the net CO₂ consumption and production due to marine silicate alteration have not been studied thoroughly. Factors such as silicate types and rates are crucial in determining the fate of CO₂ in the marine subsurface. In this study, we aimed to constrain silicate types of marine silicate alterations and their rates by measuring Si isotopic signatures of porewater and different silicate phases (including LSi, biogenic silica (BSi) and amorphous secondary Si phase (ASSi)) and modelling downcore profiles from two drill cores retrieved from the Ulleung Basin, where up to 130 mM of alkalinity has been documented as a result of fast marine silicate weathering. A decrease in porewater dissolved Si (DSi) concentration and an increase in porewater $\delta^{30}\text{Si}$ ($\delta^{30}\text{Si}_{\text{pw}}$) value indicate the formation of ASSi in the shallow subsurface (0 to 9 meter below seafloor (mbsf)). Below the sulfate-methane transition zone (9 to 32 mbsf), an increase in DSi concentrations and a decrease in $\delta^{30}\text{Si}_{\text{pw}}$ values were attributed to the LSi dissolution releasing lighter Si isotopes into porewater. Such a dissolving LSi phase is likely a mica-group silicate, as suggested by the elemental content of the separated solid phase and porewater. This finding is supported by reactive transport simulation, which indicates that mica, vermiculite and albite are able to dissolve and release Mg, K and Na into porewater. Precipitation of smectite group silicates consumes Mg and K in the pore fluids at rates lower than the overall silicate dissolution rates. The dissolving mica-group silicate (and albite) neutralises CO₂ produced through organic matter fermentation and increases porewater alkalinity, which is 60 times higher than the seawater value. By further conducting model sensitivity tests using various organic matter degradation rates, we found that the alkalinity concentrations contributing by dissolved Mg and K concentrations are majorly affected by changes in smectite group formation rates while mica and vermiculite dissolution rates remain constant. The decreased contribution of mica-like silicate dissolution and the increased contribution of BSi dissolution with sediment depth from 32 to 218 mbsf are indicated by increased DSi

concentration, increased $\delta^{30}\text{Si}_{\text{pw}}$ values and rate results output by modelling.