

Comparative geochemical properties of methane-derived authigenic carbonate (MDAC) in high methane flux areas in the southern Ulleung Basin and the ARAON Mounds in the Chukchi Sea

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We collected methane-derived authigenic carbonate (MDAC) from high methane flux areas in the southern Ulleung Basin, South Korea, and the ARAON Mounds in the Chukchi Sea during expeditions. This study compares the chemical properties of MDACs from both regions and examines how methane (CH₄) seepage influences their chemical signatures. Gas composition and isotopic signatures of CH₄ ($C_1/C_2^+ > 300$, $\delta^{13}C_{CH_4} < -60\text{‰}$, $\delta D_{CH_4} \leq -190\text{‰}$) indicate a microbial CH₄ source in the southern Ulleung Basin, whereas those in the ARAON Mounds suggest both thermogenic and microbial origin ($C_1/C_2^+ > 300$, $-82\text{‰} < \delta^{13}C_{CH_4} < -47\text{‰}$, $\delta D_{CH_4} \leq -178\text{‰}$). In the southern Ulleung Basin, MDACs exhibit diverse morphologies (massive-porous, massive-laminated, and semi-/unconsolidated) and are associated with bioclasts, planktonic foraminifers, diatoms, and radiolarians. They primarily consist of aragonite and magnesian calcite, with carbon isotopic values ($\delta^{13}C_{MDAC}$) below -31.3‰ . Higher $\delta^{13}C_{MDAC}$ values above the sulfate-methane transition zone (SMTZ) ($> -34.3\text{‰}$) suggest a combined influence of seawater and upward-migrating CH₄ on MDAC precipitation, while lower values below the SMTZ ($< -41.6\text{‰}$) indicate a dominant CH₄ influence. Additionally, vertical variations (3.9‰ – 5.8‰) in $\delta^{18}O_{MDAC}$ values, compared to theoretical value ($\sim -4.0\text{‰}$), reflect an association with gas hydrate (GH) dissociation and formation. In contrast, MDACs at the ARAON Mounds are devoid of bioclasts and mainly consist of magnesian calcite and dolomite. $\delta^{13}C_{MDAC}$ values range from -34.4‰ to -25.2‰ , indicating thermogenic CH₄ as the predominant source. $\delta^{18}O_{MDAC}$ values (4.6‰ to 6.2‰) exceed the theoretical equilibrium value, suggesting incorporation of ^{18}O -enriched fluid from GH dissociation into carbonate precipitation. Overall, the chemical signatures of MDACs from the southern Ulleung Basin and the ARAON Mounds have been significantly influenced by the impact of CH₄ migrated from deep-seated sediments, as well as