Supplementary Information

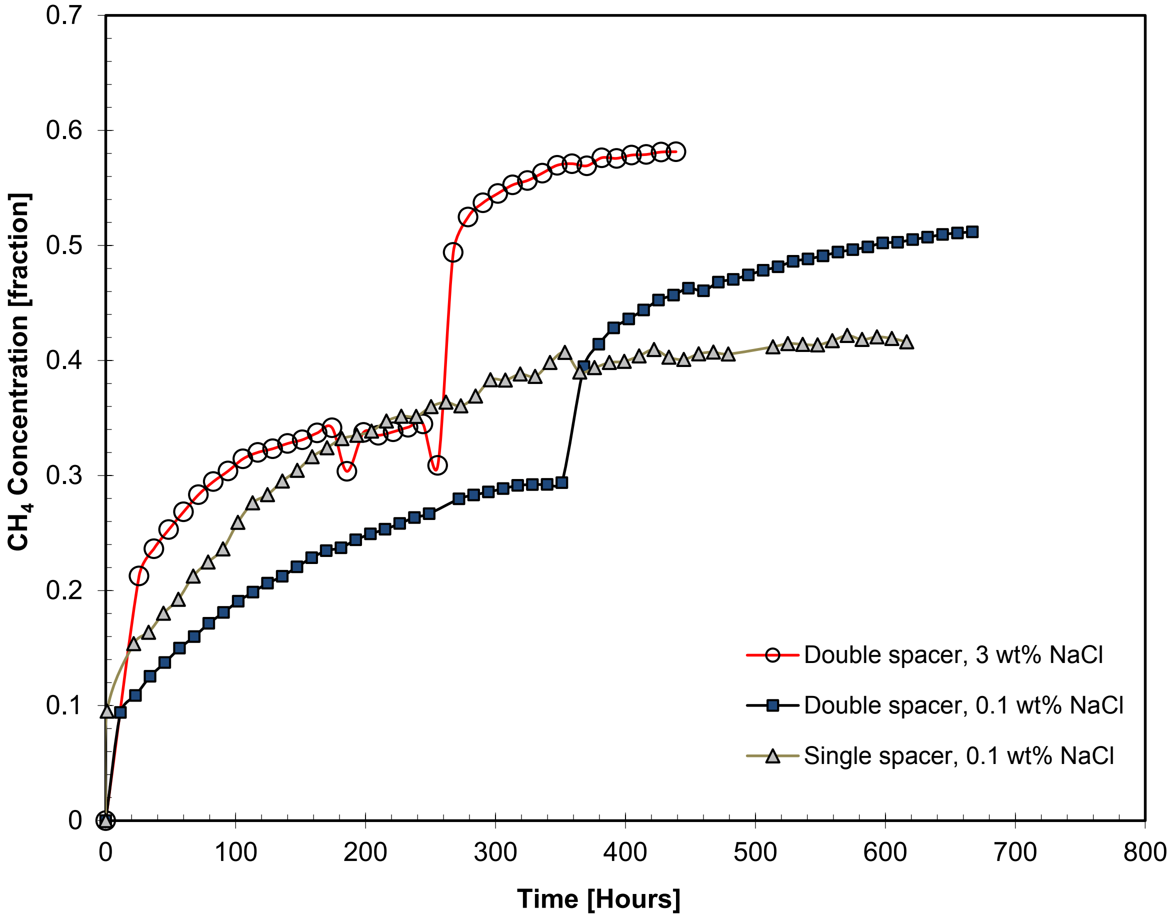

Figure S1. Comparison of CH_4 build-up in the spacer volume during CO_2-CH_4 exchange for two double spacer experiments and one single spacer experiment. Differences in sample configuration complicates direct comparison; however, similar behavior was observed for the three experiments.

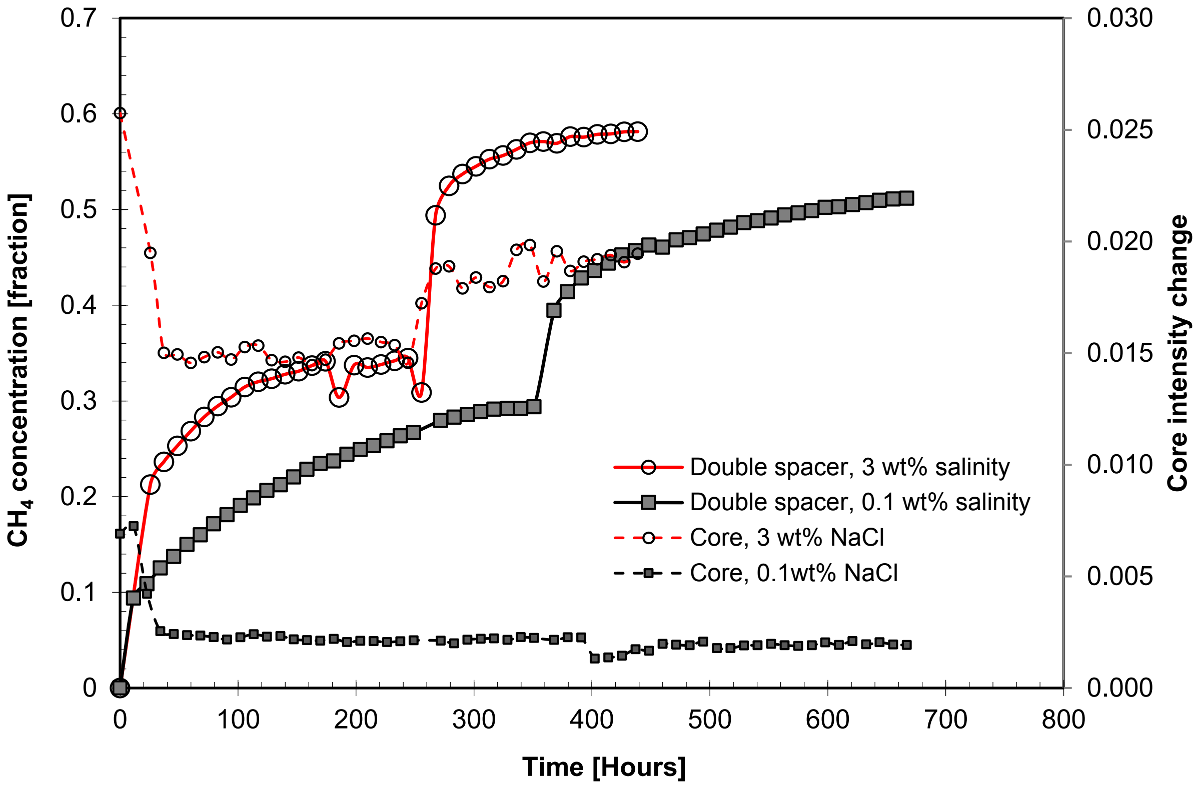

Figure S2. Comparison of intensity variations during the CO_2-CH_4 exchange process for both core segments (average of all three) and variations in the spacer volume (average of both). A significant intensity drop was observed in the core segments after the CO_2 flush, which indicates that CO_2 is transported into the core segments and reacts with the CH_4 hydrate and/or the excess water. The intensity is higher for the sample with initial 3 wt% NaCl brine, as more excess water is present at the initiation of the CO_2 flush($X_{NaCl = 3\ wt\%}$: $S_w = 0.081$, $X_{NaCl = 0.1\ wt\%}$: $S_w = 0.023$). An unexpected intensity increase was observed after the 2nd flush (after 256 h), which indicates significant CH_4 release or partial hydrate dissociation. Similar trends were not observed for the sample with lower salinity. The overall negative intensity change during CO_2 soaking suggest that the hydrate saturation (most likely stabilized by CO_2) increased throughout the experiment.

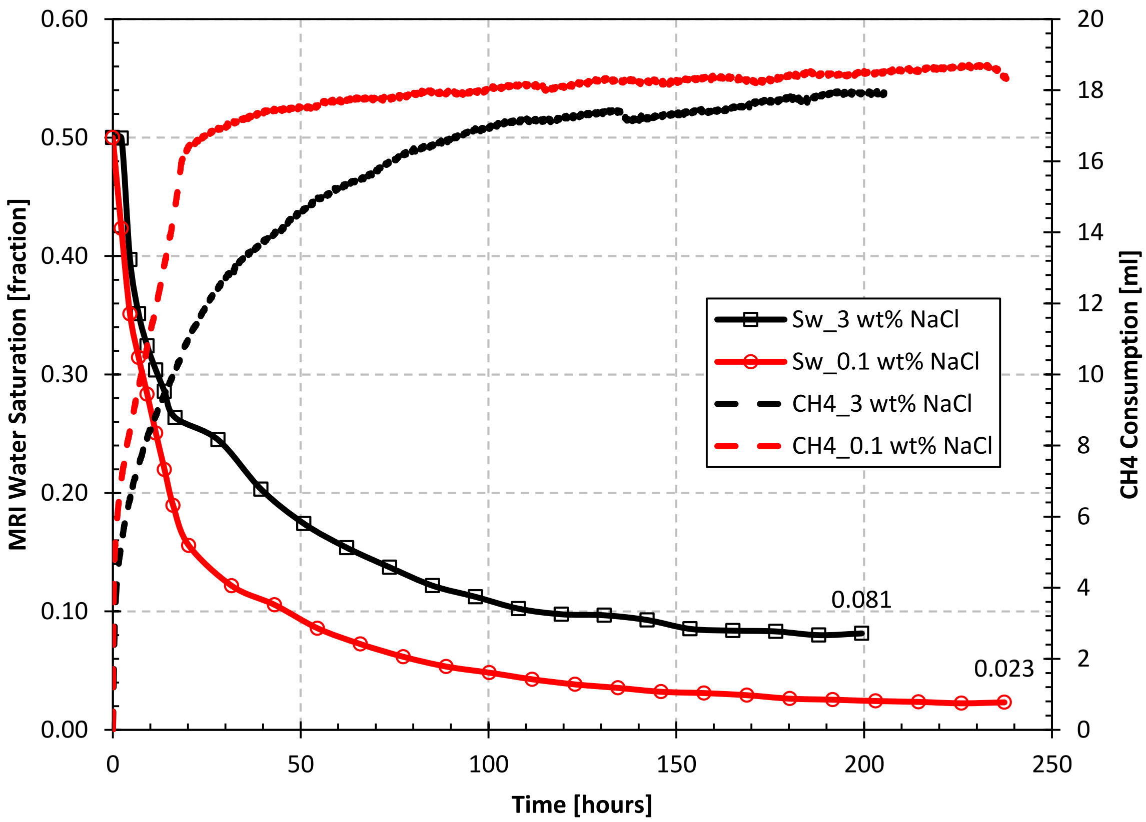

Figure S3. Comparison of water saturation (derived from MRI) and CH_4 consumption during hydrate growth for the two double spacer experiments. Higher water saturation ($S_w = 0.081$) was observed for the experiment with higher brine concentration (black curves), while low water saturation ($S_w = 0.023$) was observed for the low salinity experiment (red curves). Abundance of water may be beneficial from a mass transportation perspective due to significantly higher diffusivity.

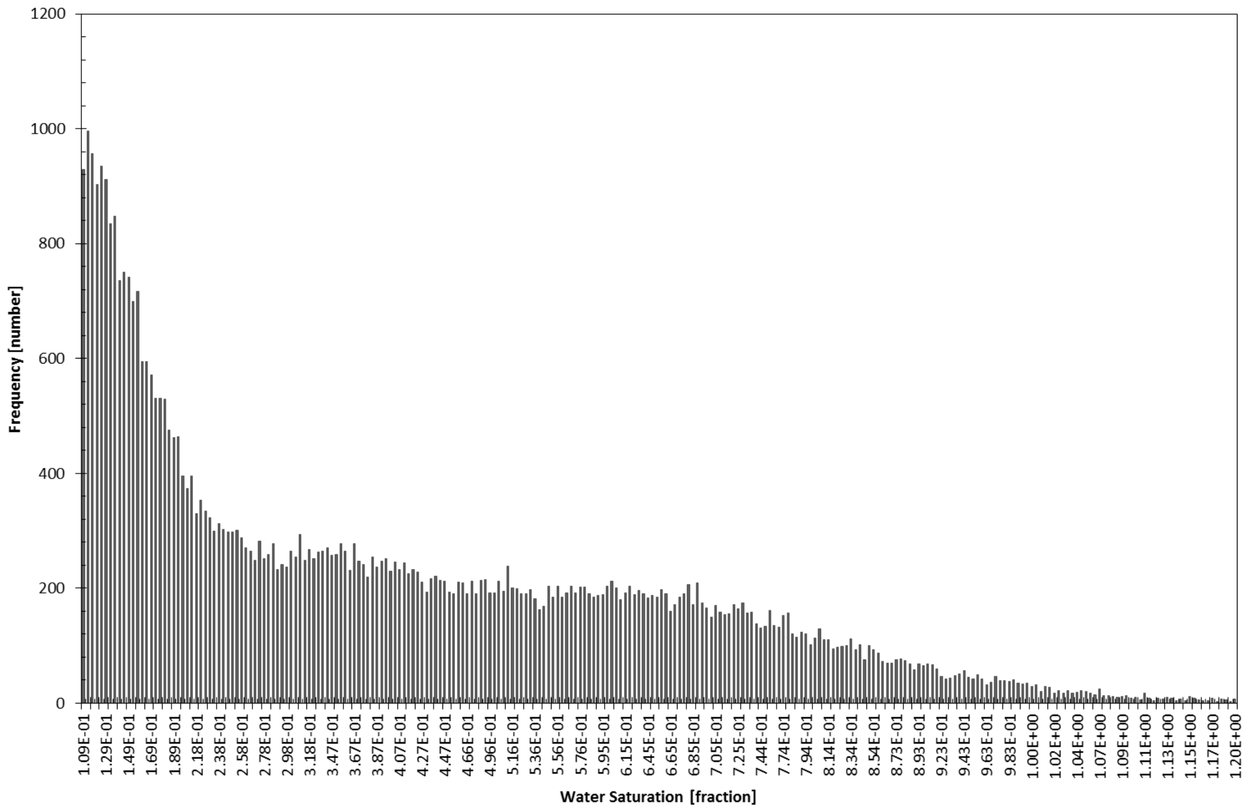

Figure S4. Initial water saturation in experiement c4 ranged between 0 and 1, where the lower values (left end of histogram) were in the background range, while the higher values are likely influenced by proximity to the open spacer volume which impacts the relaxation properties and over-estimates water saturation. Values exceeding 1 are therefore outliers, but have still been included. Mass transport through areas with high initial water saturation may become challenging as gas hydrates form and reduces the effective pore space.

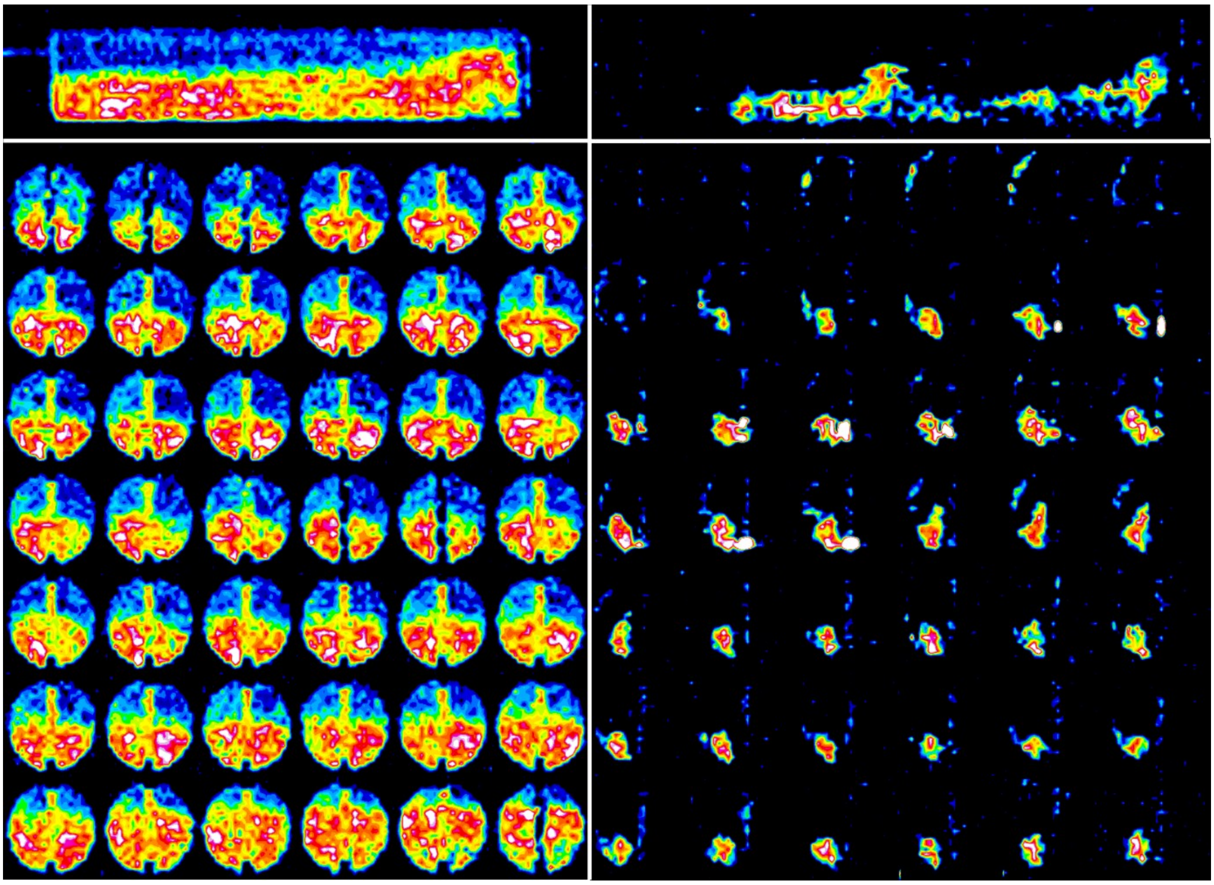

Figure S5. c4 was characterized by non-uniform initial water saturation, as illustrated in the left part of the figure. The upper images are sagittal slices of the sample, while the lower circular images show transverse slices at different positions along the core length. The bottom section of the sample was characterized by consistently higher water saturation, even exceeding 90% in local areas. The right section of the curve shows that initial dissociation during depressurization was focused in areas characterized by high initial water saturation, which suggests that the exchange process was less effective in these areas.

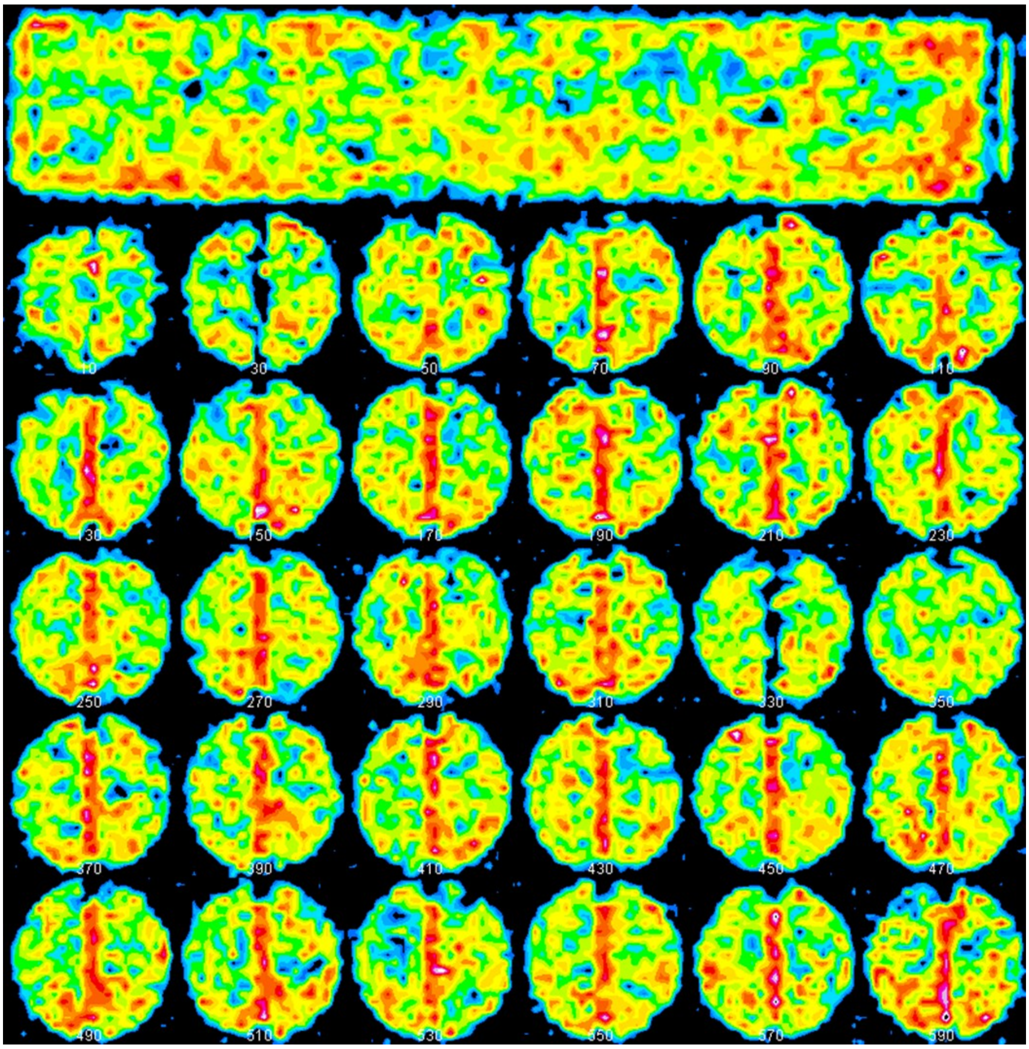

Figure S6. Gas expansion during dissociation for c4 resulted in water re-distribution and fairly uniform S_{wi} for experiment c5.