



## Exploring Carbon Sequestration through Mineral Carbonation in Serpentinite Mud Volcanoes

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Mineral carbonation is a sequence of natural processes that can be summarized as a net reaction of carbon dioxide (CO<sub>2</sub>) with metal-bearing minerals, producing stable carbonates. The rapid increase in anthropogenic CO<sub>2</sub> emissions has created an imbalance in the carbon cycle which natural mineral carbonation is not able to offset by itself. The severity of the problem now requires the use of an array of carbon sequestration techniques, such as Carbon Dioxide Capture and Storage (CCS) and Carbon Dioxide Removal (CDR) in addition to drastically reduced greenhouse gas emissions. Mineral carbonation has been drawing attention as a potentially sustainable technology to reach carbon neutrality, by storing CO<sub>2</sub> as carbonate minerals that are stable over a long period of time. Thus, many recent studies have focused on characterizing the chemical reactions that occur during mineral carbonation and developing methods to improve its efficiency, however, mostly under laboratory conditions.

The Mariana forearc presents a unique opportunity to study these processes in a natural system, as mineral carbonation occurs as authigenic carbonate precipitates in the serpentinized muds from large mud volcanoes. We analysed samples collected from three serpentinite mud volcanoes – Yinazao, Asút Tesoru and Fantangisña – during the IODP 366 expedition to characterize the C, O and Ca isotopic composition of these authigenic carbonates. They consist of rhombohedral calcite and aragonite needles and spherulites found predominantly in the core's top meters. At Yinazao aragonite occurs in the mud volcano's summit, showing a  $\delta^{13}\text{C}$  of  $\sim 0 \pm 0.9\text{‰}$ ,  $\delta^{44/40}\text{Ca}$  of  $\sim 0 \pm 0.5\text{‰}$ , and  $\delta^{18}\text{O}$   $\sim 5 \pm 0.3\text{‰}$ , while calcite is found on the mud volcano's flank, with higher values of  $\delta^{13}\text{C}$  and  $\delta^{44/40}\text{Ca}$  ( $\sim 2.9 \pm 0.08\text{‰}$  and  $1.4 \pm 0.06\text{‰}$ , respectively), and lighter  $\delta^{18}\text{O}$  ( $\sim 1.7 \pm 0.43\text{‰}$ ). As for Asút Tesoru and Fantangisña we also found aragonite needles and spherulites on the mud volcanoes' flank and summit, respectively, with similar  $\delta^{13}\text{C}$ ,  $\delta^{44/40}\text{Ca}$  and  $\delta^{18}\text{O}$  isotopic signatures compared to the ones from Yinazao.

These results emphasise the importance of these mud volcanoes as ideal models to study mineral carbonation reactions in a naturally occurring system. Importantly, they show that seawater is the

major source of carbon for the authigenic carbonate precipitation. This precipitation is the result of the reaction between seawater with the highly alkaline fluids sourced by serpentinization reactions that ascend through the mud volcanoes.

This knowledge highlights the importance of mineral carbonation as a potentially effective approach for both CDR and CCS, as well as the need for further studies in natural systems. Additionally, it shows that understanding how mineral carbonation occurs naturally may be the key to overcoming many of the challenges we currently face when developing efficient carbon sequestration technologies.