Tracing volatile cycling during progressive subduction in the Mariana Forearc

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Geological processes at subduction zone margins control seismicity, plutonism/ volcanism, and geochemical cycling between the oceans, crust, and mantle. The down-going plate experiences dehydration, and the associated metamorphism alters the physical properties of the plate interface and mantle wedge. Although volatile loss during accompanying volcanism is reasonably well quantified, there are few constraints on volatile transfer to the forearc. Consequently, it is not known how much carbon is retained in down going slabs at subduction zones and transported to the deep mantle. Here we interrogate a unique sample set of fluids and xenoliths that have ascended from the subduction channel and erupted at serpentinite mud volcanoes in the Mariana forearc to quantify the volatile budget of a subducting slab.

We make key observations of changes in pore and borehole water chemical and isotopic composition with progressive subduction; and map where key reactions take place within the subduction channel. Pore fluids from the shallowest depths-toslab (13-16 km) are Ca and Sr-enriched, but otherwise solute poor, low alkalinity fluids of pH ~11. In contrast, the deeperderived fluids (>18 km) have high pH (12.5) and elevated DIC, Na and Cl, as well as B and K, which are associated with the breakdown of slab sheet silicate phases. These fluids also are depleted in Ca and Sr compared to seawater and have higher δD and δ^{11} B values than shallower waters (δ D values up to +16 %; $\delta^{11}B \sim 14\text{-}15 \text{ \low cf. } \delta D < 0 \text{\ \low ; } \delta^{11}B \sim 12\text{-}13 \text{\ \low)}.$ These data indicate that the lawsonite-epidote mineral transformation boundary ~250 °C (>18 km depth) is an important source of devolatilization waters and may drive slab carbonate destablisation, despite its apparent thermodynamic stability at such temperatures and projected pressures (~300 °C and ~0.6 GPa). At shallower depths, the main reactions controlling fluid liberation are sediment compaction at the shallowest depths (<13 km) followed by clay diagenesis and desorbed water release (>13 km depth). These new data provide direct evidence for the

progressive mineralogical and chemical evolution of a subducting oceanic plate that is liberating progressively evolving fluid phase to the subduction channel.