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High-pressure high-temperature Raman spectroscopy of kerogen: Relevance to subducted organic carbon

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ABSTRACT

The amount of insoluble macromolecular organic matter in the Earth's crust, commonly referred to as kerogen, far exceeds the mass of living organic matter. The fraction of kerogen in sediments subducted into the mantle remains poorly constrained and will vary depending on the physical-chemical properties of kerogen along different slab geotherms. We studied the pressure-temperature evolution of carbon vibrational frequencies in isolated kerogen, previously not subjected to metamorphism, using Raman spectroscopy in a sapphire optical cell up to 3.2 GPa and 450 °C, corresponding to colder subduction geotherms. For blue-green laser excitation, we find optical irradiance exceeding ~3 kW/cm^2 induces changes in spectral features of the primary graphitic (G-band) and two main disordered modes (D1 and D2) that might otherwise be mistaken for thermal maturation. Whereas previous in situ studies have investigated the changes in these molecular vibrations of kerogen at high temperature or high pressure, we collected Raman spectra of isolated kerogen at simultaneous high P-T conditions. Although instantaneous and irreversible changes in band ratios of isolated kerogen were observed above ~350 °C at room-pressure, long-duration (2–8 h) heating experiments at 450 °C and 2.7–3.0 GPa reveal no permanent change in band structure. The reduction in vibrational frequencies of the disordered carbon modes with temperature ($d\nu/dT$) at pressures >1 GPa is slightly less than found at room pressure, further indicating that pressure effectively increases the thermal stability of kerogen. Our results suggest that kerogen reaching depths of 60 km where the temperature is below ~450 °C may subduct into the mantle, providing a potential source for the organic-rich component of carbon recently detected in certain lower-mantle diamonds.

Keywords: Kerogen, Raman spectroscopy, subduction, sapphire anvil