Title:	The reliability of Raman micro-spectroscopy in measuring the density of CO_2 mantle fluids
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Abstract

Recent evaluations of carbon fluxes into and out the Earth's interior recognize that a significant part of the total outgassing of deep Earth carbon occurs in tectonically active areas (Kelemen and Manning, 2015). Potential tracers of carbon fluxes at mantle depths include CO2 fluid inclusions in peridotites. Raman micro-spectroscopy allows calculating the density of CO2 fluids based on the distance of the CO2 Fermi doublet, Δ, in cm-1 (Rosso and Bodnar, 1995). The aim of this work is to check the reliability of Raman densimeter equations (cf. Lamadrid et al., 2016) for high-density CO2 fluids originating at mantle depths. Forty pure CO2 inclusions in peridotites (El Hierro, Canary Islands) of known density (microthermometry) have been analyzed by Raman micro-spectroscopy. In order to evaluate the influence of contaminants on the reliability of equations, 22 CO2-rich inclusions containing subordinate amounts of N2, CO, SO2 have also been studied. Raman spectrometer analytical conditions are: 532 nm laser, 80 mW emission power, T 18°C, 1800 and 600 grating, 1 accumulation x 80 sec. Daily calibration included diamond and atmosphere N2. Results suggest that the "Raman densimeter" represents an accurate method to calculate the density of CO2 mantle fluids. Equations, however, must be applied only to pure CO2 fluids, since contaminants, even in trace amounts (0.39 mol%), affect the Δ resulting in density overestimation. Present study further highlights how analytical conditions and data processing, such as spectral resolution (i.e., grating), calibration linearity, and statistical treatment of spectra, influence the accuracy and the precision of Δ measurements. As a consequence, specific analytical protocols for single Raman spectrometers should be set up in order to get reliable CO2 density data.

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