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Arsenic Distribution and Speciation in Appalachian Serpentinites in Northern Vermont, USA

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High concentrations of As in ground waters are noted in many parts of New England (Ayotte et al., 2003). Previous work by Ryan et al. (2009) documented that serpentinites in northern Vermont contain high As, up to 449 ppm. We selected representative samples of serpentinite and examined the distribution and speciation of As in the samples. The serpentinites are composed of serpentine with minor Cr-spinal, magnetite, and carbonates. The XRD spectra of the samples show that antigorite is the serpentine phase. Carbonate minerals are magnesite and dolomite with very minor calcite. Cr-spinel grains are variably altered to ferritchromite and magnetite in their rims. Samples are separated into a magnetic fraction and a non-magnetic, serpentine-rich fraction. Leaching of a serpentine-rich fraction with NaH_2PO_4 (0.1M) yielded As, which is less than 10 % of the total As recovered by hot HF/HNO_3. The data suggest that As is mostly incorporated in the crystal structure of antigorite. This is further confirmed by the quantitative analysis of minerals using an electron microprobe. Antigorite contains variable, but high concentrations of As, up to 1300 ppm. X-ray absorption spectra at As K-edge show that As is +3. Local atomic structures around As are calculated based on the EXAFS spectra. The results show that the coordination number of As is 4.0 and that the atomic distances are 1.778±0.009 Å between As and O, 3.196±0.070 Å between As and Si, and 3.4875±0.032 Å between As and Mg. The data suggest that As replaces Si in the tetrahedral site of antigorite.

Earlier study of serpentinites in north western Himalayas suggested that As is +5 replacing Si in antigorite (Hattori et al., 2005). The data from the two locations suggest that antigorite is capable to incorporate As (+3) and As (+5) into its tetrahedral site.