The fluorine and chlorine budget of the Earth's peridotite mantle

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The presence of halogens in the Earth's mantle may significantly affect its melting dynamics and rheological properties. Previous studies have provided insights into halogen partitioning during mantle melting and the halogen contents of mantle-derived melts (MORB and arcs), however the distribution of F and Cl in the most common peridotite minerals, namely olivine (ol), clinopyroxene (cpx), and orthopyroxene (opx), is still poorly constrained. Here we have developed a protocol to measure concentrations down to 0.4 µg/g for F and 0.3 µg/g for Cl in mantle minerals by secondary ion mass spectrometry. We have measured F and Cl (±Li, Be, B) in coexisting olivine and pyroxene (±amphibole, am) in twenty natural peridotite samples from various tectonic settings, including forearc peridotite (Josephine Ophiolite, USA), oceanic peridotite (16°N Mid Atlantic Ridge, and Dish Hill, CA), sub-continental lithospheric mantle (Colorado Plateau, USA), and metasomatized mantle (Finero-Balmuccia, Italy). We find that the order of fluorine compatibility ranks as follows: am>cpx>opx≥ol, while for chlorine: am>cpx>ol~opx. We show that F incorporation into pyroxene is mainly controlled by crystal chemistry and that F and Cl distribution in mantle minerals varies drastically among tectonic settings. Halogen contents could thus provide a sensitive indicator for wet versus dry melting, and for metasomatism by silicate melts versus aqueous fluids. We calculated that bulk fluorine concentrations for nominally anhydrous ol-opx-cpx upper mantle vary from 1.4 µg/g to 31 µg/g, and oceanic peridotites yield a more restricted bulk fluorine content of 2.1–9.4 µg/g F. Anhydrous minerals contain less than 1 µg/g Cl in all samples, and calculated bulk chlorine contents for an ol-opx-cpx mantle range from 0.14 to 0.38 µg/g, which is lower than estimates available for the Depleted MORB Mantle. Our results show that an ol-opx-cpx peridotite mantle alone cannot account for the uncontaminated Cl contents of MORB, and that a F-rich eclogite component may be required to explain the F content of MORB.