Sulfate in fluid inclusions of the Pinatubo harzburgite xenoliths as slab-derived oxidiser in the mantle wedge

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We found sulfate ion and sulfate minerals in the H_2O-CO_2-Cl fluid inclusions in the harzburgite xenoliths collected in the Pinatubo 1991 pyroclastic flow located at the volcanic front of the Luzon arc, the Philippines (Kawamoto et al. Proc Natl Acad Sci USA 2013). Thanks to a newly installed Raman mapping system, SO_4^{2-} ion, gypsum ($CaSO_4 \cdot 2H_2O$) and anhydrite ($CaSO_4$) were found in the fluid inclusions. Kumagai et al. (2014, Contrib Mineral Petrol) reported a possible presence of Mg-sulfate hydrite in CO_2-H_2O-Cl fluid inclusions in the Ichinomegata lherzolite xenoliths sampled in a rear arc side of northeaster Japan arc. In the previous paper at 2013, we had mentioned no sulfur phase found in the Pinatubo harzburgite, which was turned out to be wrong. Halogen systematics of the Pinatubo harzburgite xenoliths indicate that the fluid inclusions can be derived from dehydration of subducting serpentinites whose water was originally brought by sedimentary pore fluids through fractures (Kobayashi et al. 2017 Earth Planet Sci Lett). Sulfate and sulfide are present in serpentinites (Alt et al. 2012 Earth Planet Sci Lett). Our preliminary estimate of sulfur contents in the fluid inclusions (0.3 wt % as S) following methods by Binder and Keppler (2011 Earth Planet Sci lett) seems to be consistent with S contents in serpentinites, their dehydrated products and the dehydrated fluids (Alt et al. 2012). Our discovery of sulfate in the Pinatubo harzburgite may suggest slab-derived fluids contain sulfate beneath the cold forearc (<830 ˚C, Pinatubo) and the warm rear-arc (about 920 ˚C, Ichinomegata). Yoshikawa et al. (2016, Lithos) report trace element compositions in amphibole of the Pinatubo harzburgite and found enrichment of Pb, which can be transferred preferentially by the sulfate bearing fluids. Subduction zone magmatism has been characterized by its high water contents and oxidized conditions (Kelley and Cottrell 2009 Science). Presence of sulfate in the slab-derived fluids can explain the positive relation between the water contents and Fe^{3+}/Fe^{2+} ratios, and also proved insights on the presence of calc-alkaline rock series characterized by higher oxygen fugacity than tholeiitic rock series (Miyashiro 1974 Am J Sci). The present observation finds a missing link between high S concentration in arc magmas (Le Voyer 2010 J Petrol) and salfate in subducting slab including serpentinites (Alt et al. 2012) or high-pressure metamorphic rocks (Frezzotti and Ferrando 2007 PERIODICO di MINERALOGIA).

Keywords: fluid inclusion, aqueous fluid, subduction zone, calc-alkali magma, oxygen fugacity, saline fluids

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