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The first Pb paradox and the composition of the sub-continental lithospheric mantle

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The first Pb paradox stems from the observation that most analyses of terrestrial material yield substantially more radiogenic Pb isotope ratios than would be expected, compared to the Pb isotope ratios derived from chondritic material[1]. This suggests that an as yet unidentified reservoir of unradiogenic Pb may exist somewhere within the Earth's interior. Prior studies[2-4] suggest that this reservoir may at least partly reside in sub-oceanic lithospheric mantle. To date, very little attention has been paid in searching for an equivalent reservoir in the sub-continental lithospheric mantle (SCLM).

The chalcophile nature of Pb means that base-metal (Fe-Ni-Cu) sulphides possibly represent a major repository for Pb, yet the precise analysis of Pb isotopic compositions of 50 to 500 μm diameter sulphide grains recovered from the mantle has, historically, been limited to a very small number of studies (e.g. [2,3]) because of the difficulty in processing aliquots of Pb in the picogram to nanogram range. Recent development of a low blank 207Pb/204Pb double spike technique and use of a Triton Plus thermal ionisation mass spectrometer equipped with a 1013 Ohm amplifier to measure the low abundance 204Pb isotope at the Vrije Universiteit[5] now allows Pb isotope analyses of minute peridotite-hosted sulphide grains.

In this study we analysed the Pb isotope composition and Pb elemental abundance in 19 base-metal sulphides recovered from a single SCLM harzburgite xenolith from Kilbourne Hole, New Mexico, USA. In addition to partial oxidation to Fe-oxy-hydroxides, many of the sulphides had also subsequently interacted with a siliceous melt / fluid, obscuring their primary mineralogy and textures still further. Sulphide masses ranged from 17 to 171 μg, yielding Pb aliquots of 0.14 ng to 7.45 ng Pb. Blank measurements were consistent at 65 pg Pb, meaning that in the two smallest samples up to 45% of the measured Pb came from making the measurement itself. However, the mean blank contribution in the remaining samples was only 11% and can be corrected using the known isotopic composition of the blank. Pb isotope ratios obtained were ubiquitously radiogenic, plotting to the right of the 4.57 Gyr "geochron", suggesting that, in this harzburgite sample at least, despite a wide range of Pb abundances being retained in mantle base-metal sulphides, sulphides from the SCLM are unlikely to represent a solution to the first Pb paradox. However, the possibility that the radiogenic Pb signature of these sulphides is derived from the observed interaction with a siliceous melt / fluid cannot be entirely discounted.

References: [1] Allegre et al (1969) Earth Planet. Sci. Lett 5, 261-269. [2] Warren & Shirey (2012) Earth Planet. Sci. Lett. 359-360, 279-293. [3] Burton et al (2012) Nat. Geosci. 5(8), 570-573 [4] Malaviarachchi et al (2008) Nat. Geosci. 1, 859-863. [5] Klaver et al (2016) J. Anal. Atom. Spectom. 31, 171-178.