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Origins of high lithium content in geothermal fluids in a continental rift setting- case study of the Upper Rhine Graben, France

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The Upper Rhine Graben (URG) is a continental rift where geothermal fluids are exploited in Hercynian fractured crystalline basement and overlying Permian and Buntsandstein sandstones to produce heat and electricity since 2007. Geothermal fluids show high lithium concentrations (mean value of ~174 mg/l). A combined mineralogical and geochemical study of Hercynian granites and Permian and Buntsandstein sandstones, sampled from geothermal wells and analogue outcrops was carried out to investigate lithium sources and lithium enrichment processes that lead to the present lithium concentrations in brines. Exploited crystalline reservoirs consist of biotite-amphibole granite containing ~50-55 ppm Li. Natural analogue of such granites in Vosgian Mountains (Ringelbach catchment) show a larger range of Li concentrations of 40-115ppm depending of their biotite content. S-type biotite-muscovite granites are also present in Vosgian Mountains and contain higher Li concentrations (108-398ppm). In both granite types, biotite is the main lithium-carrier mineral with Li contents of ~500 ppm in biotite-amphibole granite and up to 2000 ppm in biotite-muscovite granites. The lithium concentrations of granites decrease in the matrix of fracture walls, mainly in relation with the breakdown of biotite. Li concentrations in Permian and Buntsandstein sandstones range between 11 and 164 ppm. The highest values are measured in Permian clay-rich sandstones with detrital white micas containing up to 2800 ppm Li. Petrological observations indicate that alteration of biotite and muscovite into illite is a key process controlling lithium mobilisation from crystalline and sedimentary reservoirs into geothermal fluids. Lithium mobilisation likely occurred during multiple geological stages: dismantling of Hercynian chain that originated Permian–Triassic deposits, hydrothermal activity in deep basin setting and continental rifting. Argillic hydrothermal stages in granite significantly redistributed lithium, with hydrothermal quartz acting as lithium sink. Present-day URG brines are close to equilibrium with Buntsandstein sandstone reservoirs at temperatures close to 225 ± 25°C, but not with crystalline basement rocks, highlighting the importance of sediment-basement transition in lithium enrichment. Lithium mass balance estimates confirm its mobilisation by geothermal fluids circulating through the fractured network of granitic reservoirs.