Behavior of Rare Earth Elements in spring waters: Implications for origin and upwelling process of deep brine

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Fluid generation and migration along a subducting slab has been investigated by high-pressure experiments, numerical simulations, and geophysical and geochemical observations, which have clarified that dehydration of subducting slab occurs in two steps; (i) first it occurs beneath the forearc region which hydrates the overlying mantle and then (ii) the hydrated mantle releases fluid beneath the volcanic region in subduction zones. However, there is a possibility for the fluid (i) above leaking toward the surface without being absorbed in the mantle for several potential mechanisms, such as cracking along tectonic faults, or when the slab is warm to destabilize the mantle hydrous minerals (e.g., serpentine).
The Arima-type brine has been identified as a spring water upwelling in non-volcanic region, which has a high-chlorine content, a distinct O-H isotopic ratio differing from the meteoric water line, and a high ^{3}He/^{4}He ratio indicating deep-origin (Matsubara et al., 1973). In addition to these geochemical characters, which are also clarified from the large database of deep groundwater chemistry over Japan (Takahashi et al., 2011), recent studies involving numerical modelling suggest a link between the Arima-type brine and slab-derived fluid (e.g., Kazahaya et al., 2014; Kusuda et al., 2014). The Sr-Nd-Pb isotopic ratios of a dense spring water in Arima show the same linkage, which has an overlapping signature with slab-derived fluid from the subducted Philippine Sea slab (Nakamura et al., 2015). Alternatively, chemical property and behaviour of slab-derived or deep crustal fluids have been suggested as a high-chlorine water (e.g., Kawamoto et al., 2013; Sakuma and Ichiki, 2016). These studies indicate that Arima-type brine is one of such fluid leaking toward surface from the subducting slab.
Rare earth elements (REEs) of the Arima-type brine and related waters along tectonic lines in central to southwest Japan have been investigated to assess their upwelling processes and deep-seated origins (Nakamura et al., 2014; 2015; 2016; this study). The geochemical behaviour and partitioning of REEs between solid and fluid are sensitive to temperature, volatile fugacity, and pH during the upwelling processes where these parameters are potentially variable (Ohta and Kawabe, 2000). As a result, several types of spring water have been identified. A deep brine is thought to be upwelling along tectonic lines and subsidiary faults, which are variably mixed with meteoric waters to precipitates REE-bearing minerals and evolve into spring waters with a distinct REE pattern. Rather pure meteoric waters recycling through the aquifer are widely observed in the same limited area, some of which are highly carbonated by gas with high ^{3}He/^{4}He derived from deep brine. Such carbonation enhances an interaction with the host rocks of the aquifer, which may dissolve REEs (particularly light REEs) derived from host granitic rocks.
These fluid behaviours are also distinguished by a multivariate statistical analysis (Iwamori et al., 2017) that clarified three principal components (PCs) covering ~90% of the total sample variance. The first component PC-01 corresponds to a dilution process of deep brine by meteoric, previously represented by major solute binary trends, including δ^{18}O–δD systematics; (2) PC-02, represents a precipitation process of REEs from the brine; and (3) PC-03 represents an incorporation of REEs from host rocks by carbonic acidity, although compositions of the host rocks may also have a significant impact on the spring water compositions. A comparison of the spring waters along tectonic lines has revealed a systematic geographic distribution. For instance, the deep-brines occur along the Median Tectonic Line, and the meteoric waters carbonated by the deep gas occur in the eastern part of the Kii area. The latter seems to upwell in the restricted region where deep low-frequency tremors are observed. Along the

Itoigawa-Shizuoka Tectonic Line, the variety of REE pattern is also lined to geographical distributions. Fluid chemistry would be a good indicator for deciphering the tectonic setting and/or temporal evolution of fluid upwelling.