The Loss of Iron from the 2.7 Ga Mt. Roe Paleosols (Pilbara, Australia) by Methane-rich Hydrothermal Fluids

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Previous investigators (e.g., Macfarlane et al., 1994; Rye and Holland, 1998, 2000; Yang et al., 2001) have recognized an extensively developed sericite zone in the upper part of the 2.7 Ga Mt. Roe Basalt near Whim Creek, Pilbara, Australia, which show major losses of Fe. These investigators have concluded that the losses of Fe occurred during soil formation 2.7 Ga ago; they have argued that this is strong evidence for an anoxic atmosphere prior to 2.2 Ga. However, our investigation suggest a strong possibility that the Mt. Roe paleosols formed under an oxygenated atmosphere and subsequently suffered Fe losses by methane-rich hydrothermal fluids.

The stratigraphic sequences of the Mt. Roe section, from the lower to the upper section, are consisted of: (a) the foot-wall basalt lava, (b) basaltic tuff with thin lava, (c) clastic sediments (mostly sandstones), (d) limestone with stromatolite texture, and (e) the hanging-wall basalt lava. The development of chlorite is found throughout the section. Sericite-rich rocks are generally situated on the upper part of chlorite zone. Sericite alteration occurs mainly in the basaltic tuff. The degree of sericitization is typically not correlated to that of chloritization. Many sericite bands occur in the strongly chloritized zone, as well as in the rather fresh basalt (Fig. 1). The thickness of such banding varies from ten centimeters to several meters. In places, textures showing replacement of chlorite by sericite are clearly observed, suggesting that sericitization occurred after chloritization. Even in strongly chloritized and/or sericitized rocks, the original rock textures are recognizable.

The chlorite zone are geochemically characterized by the depletion of Ti-normalized Na, Mg, Ca, Sr and Si, but not in Fe. However, sericite zone is characterized by the strong depletion in Fe, as well as in Na, Mg, Ca, Sr.

Numerous black veins, veinlets and vesicles, containing sericite, REE phosphate, cristobalite and quartz, are common. These veins are roughly concordant with the sedimentary structure, and are predominant in the sericite zone. In the most intensive veining zone, there is also another type of veins, composed of pyrophyllite, diasporite, rutile, apatite and sericite. These veins are characterized by the enrichment of so-called “insoluble elements”, such as Al, Ti, Hf, Zr and REE. That is, the soluble elements, such as alkali-, alkaline earth elements, and Fe, were almost completely leached out from the vein hosts.

Fluid inclusions in diaspore crystals in the veins show filling temperatures from 140 to 220°C and the salinity from 10-15 wt % NaCl equivalent. Some inclusions are rich in CH₄; CO₂ was not observed in any inclusions. These characteristics indicate that the hydrothermal solution were strongly reducing, which explains the losses of Fe from the paleosols. Fluid inclusions in vein quartz from cratic sediment or evaporate show the filling temperatures of ca. 115°C, providing the boiling phenomena. The inclusions in quartz are also rich in CH₄, but always poor in CO₂.

The mineralogical and geochemical data of the veins, especially the fluid inclusion data, strongly suggest the geochemistry of the sericite zone of the Mt. Roe basalt was essentially controlled by methane-rich, reducing hydrothermal fluids; the losses of Fe most likely occurred by hydrothermal fluids, rather than by rain water as previously suggested. The losses of Fe can not be interpreted by the weathering under an anoxic atmosphere. The geochemical data also suggest that the depth of the hydrothermal activity was shallow. This further suggests that the time from the eruption of footwall basalt, to the
sedimentation of tuff and clastic sediments, to evaporite formation, to soil formation and to hydrothermal alteration was short, perhaps within ~10 Ma.

The iron behavior in the chlorite zone needs other interpretation, because the total iron and ferric/ferrous ratio generally increase from the fresh footwall basalt to the chlorite-rich zone. This may suggest the soil formation, prior to the hydrothermal activity, occurred under an oxygenated atmosphere.

The sandstone and other clastic sediments contain organic matters. The H/C and N/C atomic ratios are 0.05-0.1 and 0.01-0.015, respectively. The δ13C values are -40 to -30 ‰. These values may suggest the activity of methanotrophs during the methane-bearing hydrothermal activity.


