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HIGHLIGHTS AND BREAKTHROUGHS

Regolith-hosted rare-earth elements: the phyllosilicate connection

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The analyses of all or some of the rare-earth elements (Sc, Y, and the Lanthanide Series) have been used to understand petrologic processes at many scales ranging from planetary studies to studies the earth's crust (e.g. McLennan and Taylor, 2012). Though thought to be insoluble or immobile at increased temperatures and pressures, the geochemical behavior of the rare-earth elements (REE) have varied relative to each other in many subtle ways. Specific rare-earth elements (Ce, Eu) have had distinct redox behaviors within crustal rocks. Their geochemical and mineralogic attributes have made them highly useful for many petrologic studies of the Earth's crust.

An entirely different picture of the REE's geochemical and mineralogic behavior has been evident in weathering and diagenetic systems. This general idea of the increased solubility of the REE due to weathering or diagenetic temperatures can be traced at least to the 1960s (Burkov et al. 1967). The release of REE from bedrock sources (granite and sedimentary rocks) and their precipitation as secondary REE-phases and/or REE as ion-sorbed has been described recently in the Piedmont regolith and in mined kaolin beds in the Georgia Coastal Plain (e.g. Bern et al. 2017; Cheshire et al. 2018). This increased solubility of the REE might be a paradigm shift relative to the conventional thinking about the occurrences of the REE observed from the petrologic and geochemical studies of crustal rocks.

28 The REE have been regarded recently as critical metals for important
29 technologies and products (e.g. batteries, magnets, wind turbines, LED, phosphor for
30 screens; Lucas et al. 2015). The strategic nature of the REE has stimulated further
31 thinking to understand both traditional occurrences of the REE as well as to explore for
32 new and non-traditional occurrences for the REE. The REE have been mined from
33 well-known deposits (e.g. Bayan Obo, China; Fan et al., 2016; Mountain Pass,
34 California). Examples of recently studied nontraditional and sizeable occurrences of the
35 REE included: the occurrences REE as diagenetically formed REE-phosphate phases
36 in kaolin; the presence of REE in kaolin mine waste; and the sizeable amounts of the
37 REE in coal, coal fly ash, heavy mineral sand deposits, alluvial sediments, and in deep
38 sea sediments (e.g. Seredin and Dai, 2012; Hower et al., 2016; Bern et al., 2016;
39 Cheshire et al., 2018; Elliott et al., 2018; Shah et al., 2018; Van Gosen and Ellefsen,
40 2018; Takaya et al., 2018; Liu et al., 2019).

41 The REE-enriched fractions of these heavy mineral sand deposits in southern
42 Georgia and the resumption of REE mining at Mountain Pass are currently the only
43 domestic sources of the REE being mined in US. Consequently, the US is highly
44 dependent on international sources for the REE, particularly the HREE (Gd, Lu) used in
45 many technologies, materials and other industrial uses and materials. Considerable
46 amounts of the HREE are being mined from the extraction of HREE ion-sorbed regolith
47 in SE China. The ease of extraction makes these deposits highly attractive as sources
48 of the HREE. The ion-sorbed HREE accounts for approximately 70% and more of the
49 current supplies of the HREE. A growing body of work is documenting the solubility, and
50 the adsorption of these seemingly insoluble REE in sedimentary rocks, regolith, and low

51 temperature regimes. Further study of their solubility will lead to the increased
52 development of these non-traditional or novel deposits of the REE in
53 sedimentary/regolith/soil systems.

54 Given the foregoing ideas regarding the increased solubility of the REE and their
55 formation as ion-sorbed REE or formation as secondary REE-minerals in sedimentary
56 and regolith settings, the paper entitled *The role of clay minerals in forming regolith*
57 *hosted heavy rare-earth deposits* by Martin Yan Hei Li and Mei-Fu Zhou is a most
58 welcomed contribution for many reasons. Li and Zhou took on the formidable task of
59 trying to understand the role of clay minerals in adsorbing REE in the regolith-hosted
60 REE deposits in SE China. Briefly, Li and Zhou found the halloysite-rich lower regolith
61 adsorbed the REE and heavy rare-earth elements (HREE) weathered from the parent
62 granite. These REE were later desorbed as the halloysite phases were transformed to
63 kaolinite in the upper regolith. These released REE migrated or translocated to the
64 deeper regolith containing halloysite. These combined mineralogical, geochemical, and
65 the pedogenic processes forming this regolith were thought to have created the large
66 concentrations of REE whose total REE on average is 2,500 ppm.

67 The sorption of metal ions at the interlayer sites of phyllosilicate minerals having
68 high layer charge is known well. However, the sorption of REE by halloysite stressed
69 the importance of edge/surface sorption processes and high specific surface area of the
70 individual grains. These new results on regolith-hosted REE should stimulate further
71 thinking and investigations regarding the about metal-phyllosilicate mineral sorption
72 processes at the atom scale. Li and Zhou further noted that compounds forming stable

73 complexes with halloysite would point to more environmentally friendly extraction
74 processes to recover the REE from these clays.

75 Finally, this paper was also a welcomed contribution in that the authors provided
76 a useful review of the site description to these regolith-hosted REE deposits. They
77 noted the occurrences of ion-sorbed REE elsewhere in south China and other tropical
78 settings (SE Asia, Madagascar, Malawi and Brazil). More interesting and germane to
79 the *American Mineralogist* readership, the authors stressed the phyllosilicate mineralogy
80 connection for understanding the ion-sorption of these seemingly insoluble REE. Li and
81 Zhou's results should stimulate further thought on the release and sorption of these
82 seemingly insoluble REE onto clay particles at the atom or nano-scale. This new model
83 will be useful to find further REE prospects in regolith settings.

84 **Acknowledgement**

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87 and beach sands in southern Georgia.

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