Analysis of water movement through an unsaturated soil zone in a volcanic island using oxygen and hydrogen isotopes

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In order to analyze water movement through an unsaturated soil zone in Jeju Island, soil waters were collected biweekly at three different soil depths using suction lysimeters. For comparison, precipitation was collected monthly using a plastic bottle. Temporal variations in the oxygen-18 and deuterium isotopes of the precipitation and soil waters were then monitored for about one year. The stable oxygen and hydrogen isotopic compositions of the soil waters are plotted between or near the two local meteoric water lines for summer and winter precipitation indicating that the soil waters were recharged from the year-around precipitation, and evaporation was negligible even during hot summer season in the study area. Finally, mean residence times of the soil waters were estimated from the $\delta^{18}O$, δD, and deuterium excess or d-values of the precipitation and soil waters using two well-mixed or exponential models (EM) and one exponential piston-flow model (EPM). The regression results show that the combination of the exponential piston-flow model and the d-values of the precipitation and soil waters gives the most reasonable estimation of the mean residence times of the soil waters in the study area: 74 days (2.5 months) and 198 days (6.6 months) to depths of 30 cm and 60~80 cm, respectively, in the unsaturated soil zone from the ground surface. The estimated low moisture contents and hydraulic diffusivities of the soils at the three different depths also suggest that the boundary between the upper fine-grained soil and the lower coarse-grained soil may act as a capillary barrier, and the soil waters probably flow slowly through micropores rather than rapidly through macropores in the unsaturated soil zone.

Revealing the composition and crystallinity of weathered alkali feldspar surfaces by XPS, FIB and TEM techniques

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How does the composition and structure of silicate mineral surfaces change during their reaction with acidic solutions in the field? Recently, Zhu *et al.* 2006 (*Geochimica et Cosmochimica Acta* 70, 4600-4616) have described ~10 nm thick amorphous layers on naturally weathered alkali feldspars, which supports the experimentally derived ‘leached layer’ model for silicate dissolution. We have tested this model further by comparing the chemical composition and crystallinity of the near-surface regions of alkali feldspars used in pH1 laboratory dissolution experiments with minerals from the same parent rock that had been naturally weathered. Angle-resolved X-ray photoelectron spectroscopy (AR-XPS) provided the compositional data whereas information on crystallinity and structure was obtained by cutting cross-sections of grain surfaces using the focused ion beam (FIB) instrument for high-resolution TEM imaging.

AR-XPS data confirm that the outermost ~9 nm of alkali feldspars reacted with pH 1 HCl are non-stoichiometric (i.e. enriched in Si and O relative to Al and alkalis). Analyses acquired at the shallowest take-off angle are most Si-rich, indicating that the ‘layer’ is likely to be considerably thinner than the maximum ~9 nm XPS sampling depth. TEM images of FIB-produced cross-sections of the etched grain surfaces show that they are crystalline throughout; if the non-stoichiometric layer is amorphous it must be thinner than can be resolved by the FIB-TEM technique (i.e. <~2.5 nm), or if thicker than 2.5 nm it must be very irregularly distributed over the grain surface and so has not been sampled.

AR-XPS data from naturally weathered grain surfaces are more difficult to interpret. Most analyses show enrichment in Al and depletion in Si relative to the unweathered grain interior, with data for the alkalis showing no clear trend. These data are inconsistent with the presence of a silica-rich layer equivalent to that formed experimentally but may indicate that aluminosilicate weathering products have been produced. However, secondary minerals could not be found by SEM, and FIB-TEM work has again demonstrated that the feldspars are crystalline to within <~2.5 nm of their outer surface. These results indicate that any chemical and structural modification is restricted to the outermost <2 nm of these alkali feldspars and so thick ‘leached layers’ are absent.