



Primary weathering rates, water transit times and concentration-discharge relations: A theoretical analysis for the critical zone

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The permeability architecture of the critical zone exerts a major influence on the hydrogeochemistry of the critical zone. Water flowpath dynamics drive the spatio-temporal pattern of geochemical evolution and resulting streamflow concentration-discharge (C-Q) relation, but these flowpaths are complex and difficult to map quantitatively. Here, we couple a new integrated flow and particle tracking transport model with a general reversible Transition-State-Theory style dissolution rate-law to explore theoretically how C-Q relations and concentration in the critical zone respond to decline in saturated hydraulic conductivity (K_s) with soil depth. We do this for a range of flow rates and mineral reaction kinetics.

Our results show that for minerals with a high ratio of equilibrium concentration to intrinsic weathering rate, vertical heterogeneity in K_s enhances the gradient of weathering-derived solute concentration in the critical zone and strengthens the inverse stream C-Q relation. As the ratio of equilibrium concentration to intrinsic weathering rate decreases, the spatial distribution of concentration in the critical zone becomes more uniform for a wide range of flow rates, and stream C-Q relation approaches chemostatic behaviour, regardless of the degree of vertical heterogeneity in K_s . These findings suggest that the transport-controlled mechanisms in the hillslope can lead to chemostatic C-Q relations in the stream while the hillslope surface reaction-controlled mechanisms are associated with an inverse stream C-Q relation. In addition, as the ratio of equilibrium concentration to intrinsic weathering rate decreases, the concentration in the critical zone and stream become less dependent on groundwater age (or transit time)