

Mike Leeder

Second Edition

Sedimentology and Sedimentary Basins

From Turbulence to Tectonics



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From Turbulence to Tectonics

2nd Edition

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Preface

*World is crazier and more of it than
we think, Incurrigibly plural.*

Louis MacNeice, 'Snow', *Collected Poems*, Faber

The predecessors to this book, *Sedimentology: Process and Product* (Allen and Unwin, 1982) and *Sedimentology and Sedimentary Basins: from Turbulence to Tectonics* (Blackwell Science 1999) are out of print and partly outdated respectively. I have received much feedback from many persons who have used these books over the years and the current version is intended to try to recapture the spirit of a dynamic and widely applied science. Reasons of space have prevented me from dealing with the subjects of diagenesis and the transformation of sediment to sedimentary rock. I have replaced these with chapters linking sedimentology to climate, sea-level change, tectonics, sedimentary basin architecture and their role in solving interdisciplinary problems. I feel somewhat uneasy about the omission, but it strikes me that the subject of diagenesis has become so based upon the physics of subsurface water flow and the chemistry of low temperature water-rock interactions that the difference of emphasis is too much to encompass within the present text.

Progress over the past decade has been breathtaking. Take some examples: the flow dynamics of opaque mud suspensions can now be monitored by acoustic Doppler probes; knowledge of deep-sea environments has been revolutionized by improved sea-bed imaging; sedimentological reactions to climatic and sea-level change have proved robust and sedimentology contributes vitally to the understanding of the evolution of sedimentary basins, from the birth, life and death of bounding faults to the climatic and palaeontological record contained within them. Further, carbonate sediments and their contained O₂ stable isotopes play a key role in establishing ancient oceanic composition (evaporite

fluid inclusions), the palaeoaltimetry of high mountains and plateau (calcsols) and the determination of ancient climate (speleothem). All this means that sedimentology is not something that can be done in isolation; the holistic approach is that which I have taken in this book, one based on a thorough understanding of modern processes that I trust will propel the reader into an enthusiasm for the subject and a sense of its place in the wider scheme of earth sciences, specifically in attempts to read the magnificent rock record.

Who do I expect to be reading this book? You will have completed an introductory course in general geology, earth or environmental sciences, and perhaps a more specific basic one in sedimentology or sedimentary geology. You will thus know the basic sediment and sedimentary rock types and also know something of the place of the subject within the broader earth and environmental sciences. You will have enough basic science background to understand, if not feel exactly on top of, Newton's laws, basic thermodynamics and aqueous chemistry. Though mathematically challenged, like many earth scientists including myself, you should at least know where to find out how to manipulate equations to a reducible form. I make no apology for spending a little more time with basic fluid dynamics than with the thermodynamics. This is *not* because I find one more interesting or important than the other—it's just that most high school leavers and graduating university students (even those of physics) do little in the way of fluid mechanics in their syllabi nowadays and it seemed that the theme of 'sedimentological fluid dynamics' is just such a place to set up some sort of foundation. Philosophically you should want

to reduce the complicated natural world to an orderly scheme, but at the same time not want to miss out on the romance and poetry of an unclassifiable subject. You will be someone who enjoys talking and arguing with a variety of other earth science specialists.

Just a few final notes are in order.

- More involved derivations of essential physical and chemical concepts are to be found in the end section labelled ‘Cookies’. These are meant to be helpful for intellectual health. There is also a short mathematical refresher appendix. I would appreciate it if readers let me know of any mistakes or symbol typos I may have made as I hope to live long enough to make another edition, someday.
- I have tried my best to reference major developments correctly at the end of chapters, to make sure the source of specific case histories can be traced and to respect historical precedence and discoveries. References are given in abbreviated form, but quite sufficient to be of full use in rapid web-based search vehicles like *Web of Science*.
- Many graphs with data points have been generalized to ‘clouds’ or ‘envelopes’ of data points—if you wish to get the original data, go back to the cited references.

Thanks to colleagues and friends who either directly, through conversation, or indirectly through me reading their works, have inspired my continued interest in sedimentology and its many applications. I would like in particular to thank long-time collaborators and friends Jan Alexander, Julian Andrews, Jim Best, John Bridge, Rob Gawthorpe and Greg Mack for keeping my mind stretched over past years. I also extend my heartfelt thanks to former faculty colleagues at the fine Universities of Leeds and East Anglia where I have spent my professional life, together with my ex-undergraduate and graduate students, for keeping me on my toes. I am grateful to Dr Jenny Mason for writing the sections on terrestrial carbonates (section 2.9), the role of speleothems in palaeoclimatic studies (section 23.18) and, with Dr James Hodson, for compiling the reference list. Finally, thanks to the whole production team at Wiley-Blackwell and to Ian Francis for his gentlemanly encouragement to complete this project and for putting up with some delays over the past 4 years as I periodically got on with my research and real life instead!

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Brooke, Norwich
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- Cookie 8 Fig. 3.
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MAKING SEDIMENT

...the soil which has kept breaking away from the high lands during these ages and these disasters, forms no pile of sediment worth mentioning, as in other regions, but keeps sliding away ceaselessly and disappearing in the deep.

Plato, *Critias*, Vol. 9, Loeb Classical Library

Introduction

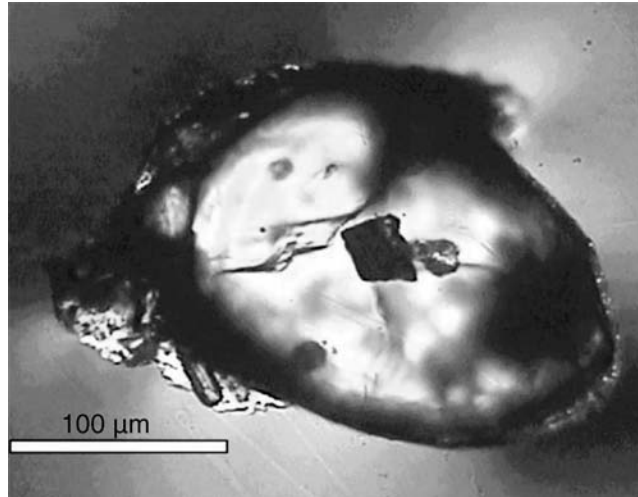
The noun *sediment* comes to the English language from the Latin root *sedimentum*, meaning settling or sinking down, a form of the verb *sedere*, to sit or settle. In earth and environmental sciences, sediment has a wide context that includes many forms of organic and mineral matter. In Part 1 we look more deeply at the origins of the sediment that occurs on and under the surface of the solid planets and which may be used to infer past environmental conditions and changes. Sediment accumulations may be grandly viewed as the great stratal archive of past surface environments, or more basically as ‘dirt’. There has been sediment on the surface of the Earth since the Archaean, with the oldest known sediment grains dating from at least 4.4 Ga (Part 1 Fig. 1). Sediment also mantles the surface of many other planets and their satellites, notably Mars, Venus and Saturn’s moon, Titan.

In scientific usage, Earth’s sediment is best divided into three end-members:

- *clastic*—originating from pre-existing rock outside a depositional area as transported grains, the commonest being mineral silicate grains, known widely as *siliciclastic sediment*;
- *chemical*—being the result of inorganic or organically mediated chemical precipitation within the depositional area;
- *biological*—derived from skeletal material associated with living tissues.

These simple divisions are robust enough to include even the highly esoteric sediment forms that are turning up in the wider Solar System, like the solid ice particles transported and deposited by liquid methane on Titan. Of course there are unusual, hybrid or mixed origins for some sediment but these can easily be accommodated (e.g. *bioclastic*, *volcaniclastic*). Note that the classification is restricted to grains that were sedimented; there are sedimentary horizons in the stratigraphic record that originated as precipitates below the deposited sediment surface, often bacterially controlled. These were never sedimented as such and are considered as secondary or *diagenetic* sediments that post-dated physical deposition of host primary sediment. Deposited sediment accumulates as successive layers, termed *strata*, and such deposits as a whole are said to be *stratified*. The succession of strata in any given deposit is controlled by environmental factors and their correct interpretation involves a deep understanding of how present and past environments have evolved over time.

The chemical and biochemical processes that produce sediment also give other soluble byproducts; these chemical species control oceanic and atmospheric composition and provide long-term sourcing for base cations that nourish plant life and counteract acid deposition in temperate forested catchments. Chemical earth-surface processes have undoubtedly changed over deep geological time, in response to atmospheric and hydrological changes, whilst biological processes



Part 1 Fig. 1 Image of the famous Jack Hills' zircons from the Cryptic Era of the Hadean Eon (discovery paper of Wilde *et al.*, 2001). The location is ~800 km north of Perth in Western Australia in the Narryer Gneiss Terrane of the Yilgarn Craton, a group of folded and metamorphosed supracrustal rocks thought to have originally contained sedimentary siliciclastic rocks. Detrital zircons with ages greater than 4 billion years old occur here, the oldest being 4.4 Ga (± 8 million), the oldest dated material originating on Earth. The source is a metamorphosed conglomerate considered to have an age ~ 3.0 Ga, so the detrital zircons are sourced from pre-existing rocks, probably subduction zone plutonic igneous intrusions which were then weathered and the resultant sediment deposited as sedimentary rock. The zircons are evidence for the existence of continental-type crust on the surface of the Earth during the Hadean Eon, contrasting with earlier ideas on the earliest phase of Earth's history in which continental crust was thought absent and plate tectonics inoperative until much later. Additionally, oxygen isotopic ratios in the zircons provide evidence for the presence of liquid water on the Earth's surface at this time. The image is a general photograph of a pristine Jack Hills' zircon (Curtin University website). Longest axis of crystal $\sim 250 \mu\text{m}$.

have changed hand-in-hand with organic evolution. By this view, sediment production is an accident of weathering and evolution—a waste product. Ever since the Archaean, the planet has 'learnt' how to cope with this waste, just like it has with the waste oxygen produced during plant photosynthesis. There was no predeterminism associated with the processes of sediment production on early Earth or any other planet. Sediment simply fell out (forgive the pun) of the rock cycle in which primary rock is chemically and physically altered. Compare for example sediment on the Moon with the Earth. In the former the sediment is

a fragmented remnant from past meteoritic impact events. In the latter sediment is highly varied in its origins, composition, size and physical properties. Its role as an accidental part of the Rock Cycle establishes sedimentology as a fundamental part of Earth System Science. Indeed, as one nice semi-popular review entitled it a decade ago (Stanley & Hardie, 1999), from the point-of-view of calcareous sediment, '*Hypercalcification: Paleontology Links Plate Tectonics and Geochemistry to Sedimentology*'. We shall examine such grand claims later in this book (Chapter 23).

CLASTIC SEDIMENT AS A CHEMICAL AND PHYSICAL BREAKDOWN PRODUCT

*Few ken to whom this muckle monument stands,
Some general or admiral I've nae doot,
On the hill-top whaur weather lang syne
Has blotted its inscribed palaver oot.*

Hugh MacDiarmid, 'The Monument', 1936, *Complete Poems*, Vol. 1, Carcanet, 1993.

1.1 Introduction: clastic sediments— 'accidents' of weathering

Terrestrial clastic sedimentary rocks are usually quite different in their composition from the igneous and metamorphic rocks that sourced them. This is because they are derived from an altered *regolith* with a soil profile produced by chemical weathering of pristine bedrock and the source of mineral grains for such sediment. For example, feldspar is the commonest mineral in bedrock of the Earth's continental crust (about 60% of the total) but quartz is usually predominant in clastic sediments and sedimentary rocks. Despite this difference the principle of conservation of mass tells us that for all elements present in the exposed crust and released by weathering, exactly the same levels of abundance must occur in the average total sedimentary mass. Thus the average *chemical* composition of *all* sediments is roughly that of the igneous rock, granodiorite, representing the mean composition of middle to upper continental crust.

It is traditional to divide rock weathering into physical and chemical components, but in reality the two are inextricably interlinked. Water is the chief reactant and plays a dual role since it also transports away both dissolved and solid weathering products. Earth is presently unique in its abundance of water and water vapour, yet Mars also had an earlier watery pre-history. It is easy to take water for granted, the deceptively simple molecule H₂O has remarkable properties of great importance for rock and mineral weathering (Cookie 1). These include its solvent and hydration properties, wetting effects due to high saturation and anomalous decrease of density at low liquid temperatures and after freezing. An outline of the near-surface terrestrial hydrological cycle is given in Fig. 1.1.

Want to know more about the structure and properties of water? Turn to Cookie 1.

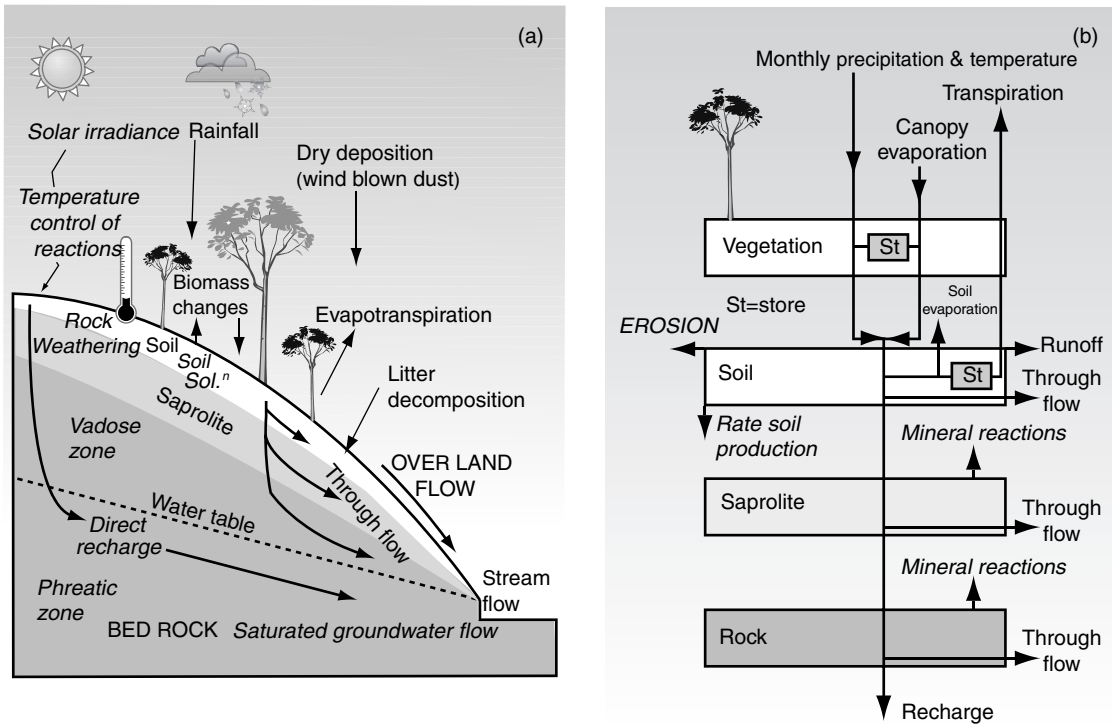


Fig. 1.1 (a) The main components of the terrestrial hillslope hydrological cycle. (b) Box model for water budget in the surface and near-surface weathering zones.

Chemical weathering involves aqueous reactions with a strong biochemical component since dissolved atmospheric gases are aided by soil-generated gases, dilute acids and organic ligands. Further, the reactions are complex since silicate minerals are involved, with their many constituent anions and cations; also the amount of water and dissolved ions varies in both time and space. Four main mechanisms contribute to chemical weathering: *dissolution*, *oxidation*, *hydrolysis* and *acid hydrolysis*. Reactions usually occur at mineral surfaces in the unsaturated (vadose) zone where, close to the local Earth's surface, rock pores contain atmospheric gases, water, living and dead vegetation and bacteria—all play an important role in weathering. The result is a regolith and soil profile whose characteristics depend upon climate and rock type.

Physical weathering involves the application of differential stresses to rock and mineral discontinuities in the unsaturated zone. These cause fragmenta-

tion and are due to erosional unloading, gravity, wind shear, salt crystallization from groundwaters, freeze–thaw and differential thermal expansion.

The combined effects of biochemical and physical weathering produce a weathered regolith profile in bedrock that comprises:

- surface *soil* with significant living and dead organic content;
- chemically altered rock termed *saprolite* that nevertheless preserves its framework coherency without volume loss (*isovolumetric weathering*);
- chemically unaltered but often physically fragmented (exfoliated) *bedrock*.

Interfaces between these layers are in a state of slow downward motion as the landscape reduces. In fact, landscape dating by *cosmogenic isotopes* and other means reveals that a steady-state system often exists, with the material mass removed by erosion being replaced by an equal volume made available from below for further decomposition.

Weathering acts on:

- mineral aggregates at or close to Earth's surface with their many intracrystalline pores and fracture networks;
 - imperfect crystals with surface and lattice defects.
- Weathering involves:
- bond *breaking*, physically by cracking and chemically by solution (see **Cookie 2**);
 - broken bond (danglers) *adoption* by ionic predators;
 - electron *removal* from the easily stripped transition metals like Fe and Mn (see **Cookie 3**).

Weathering depends on:

- H₂O *throughput* or hydraulic conductivity by laminar water flow in pore spaces between minerals, microporosity within minerals and cracks across minerals;
- H₂O *dissociation* into the reactive H and OH ions (see **Cookie 1**);
- CO₂ *concentration* via atmospheric and soil processes;
- temperature, which controls both *reaction rates* (Fig. 1.2) in silicate minerals via the results of the *Arrhenius principle* (see **Cookie 3** and further

below) and the rate of aqueous throughput via its control on water's *dynamic viscosity*.

Want to know more about ions, electron transfer and Eh-pH diagrams? Turn to **Cookie 2**.

Want to know more about proton donors, pH, acid hydrolysis and calcium carbonate weathering? Turn to **Cookie 3**.

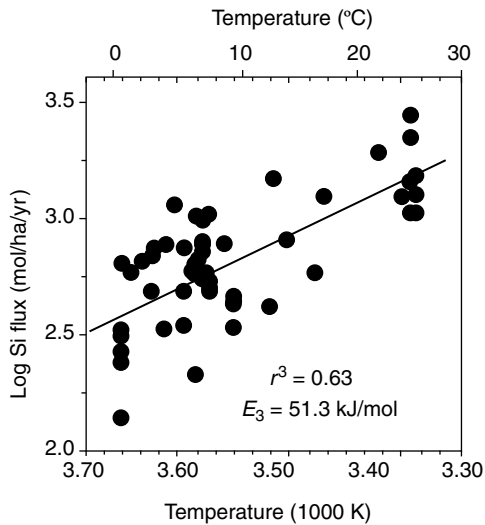


Fig. 1.2 Arrhenius plot to show that the flux of Si from small, simple-geology granite catchments depends on temperature, once corrections are made to allow for rainfall amount. This is because the rate constant is determined by the Arrhenius effect discussed in the main text and **Cookie 3**. The solid diagonal line corresponds to the activation energy E_a . (After White *et al.*, 1999.)

1.2 Silicate minerals and chemical weathering

The flux of dissolved elements and altered minerals from continent to ocean is largely controlled by the processes of chemical weathering. Global river sampling indicates that silicate weathering accounts for ~45% of total dissolved load, calcium carbonate weathering ~38% and evaporitic salts ~17%. The recharge of dissolved ions into rivers from the weathering zone (Fig. 1.3) is not only a function of source rock but also of the fluxes from atmospheric deposition, vegetation growth and respiration, and the net reaction of soil and other subsurface interstitial water with minerals in the unsaturated zone. Once liberated by weathering into the hydrological system, some elements behave *conservatively* in that they then proceed down-catchment with little further gain or loss. Other minerals behave in a decidedly *non-conservative* way —Ca is a good example since it readily forms mineral precipitates in semiarid soils and in other terrestrial carbonate sinks (section 2.9).

Chemical weathering of silicate minerals plays a major role in the global hydrogeochemical cycle. Since the relative proportion of minerals in clastic sedimentary rocks is different from those in igneous and metamorphic rocks, it is clear that some are more stable than others in the weathering process and some are newly formed; this depends upon the thermodynamics of the reactions involved (**Cookie 4**). Reactions in natural waters include rapid dissolution of ionically bonded minerals like soluble salts (**Cookie 1**) and acid attack on carbonate minerals (**Cookie 2**). Both carbonates and evaporites usually dissolve *congruently*,

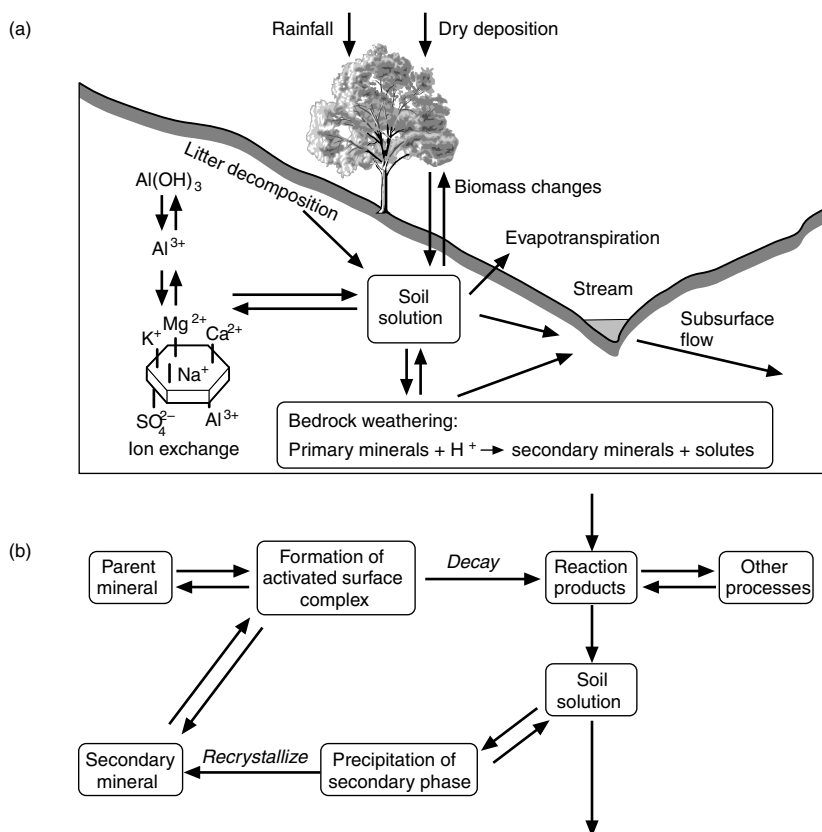


Fig. 1.3 (a) Some of the processes affecting solute fluxes in catchments. (After Drever & Clow, 1995.) (b) Flow diagram to show characteristics of surface mineral reactions. Note that incongruent behaviour involving mineral precipitation is characteristic of many weathering reactions. Also, non-conservative behaviour means that some dissolved ions may be precipitated *in situ* as other mineral phases, which therefore do not stay in solution to be exported by river water. (After Sverdrup & Warfvinge, 1995.)

i.e. the proportion of elements in solution is the same as the proportion in the former mineral(s). However, in the context of the whole catchment some of the dissolved ions may subsequently become involved in secondary mineral precipitation.

Want to know more about the thermodynamics of weathering reactions? Turn to Cookie 4.

Decomposition of silicate minerals is always *incongruent*—the reactions are very slow and temperature dependent, yielding other solid pro-

ducts in addition to dissolved ones. Reaction pathways vary according to local Eh–pH–temperature conditions. They proceed predominantly by *acid hydrolysis* in which activated surface complexes play a major role (Fig. 1.3) and where small, highly charged protons displace metallic cations in crystal silicate and oxide lattices. Silicic acid is produced from the intermediate metal-bonded silicate complexes and released to solution. OH[−] or HCO₃[−] ions finally combine with displaced cations to form solutions or local precipitates. Hydrolysis acts along lattice surfaces exposed by discontinuities such as joints, rock cleavage, crystal boundaries, mineral

cleavage planes and crystal surface defect sites. Once a dissolved ion is liberated from a crystal lattice by hydrolysis, it may link up with surrounding water molecules and thus stay in solution as a hydrated ion, or, if the force of attraction with water is insufficient to compete with that between the water molecules themselves, the ion will be 'ignored' by the water and will precipitate. Some elements will be exported from the weathering site, perhaps far away. Others will behave non-conservatively and precipitate locally, perhaps forming economic accumulations like in the concentration of Al and Fe in

bauxites and *laterites* respectively. The type of behaviour in solution is determined by the *ionic potential*, which is the ratio of ionic charge to ionic radius (Fig. 1.4). Cations (metals) whose potential is less than 3 are easily hydrated and highly mobile; anions of potential >12 form soluble complexes. Ions with potentials between 3 and 12 are mostly precipitated as hydroxides (in the absence of protons) and are thus immobile.

The proton concentration of soil water gives rise to pH values in the range 5–9, in which silicon is more soluble than aluminium. Thus prolonged chemical

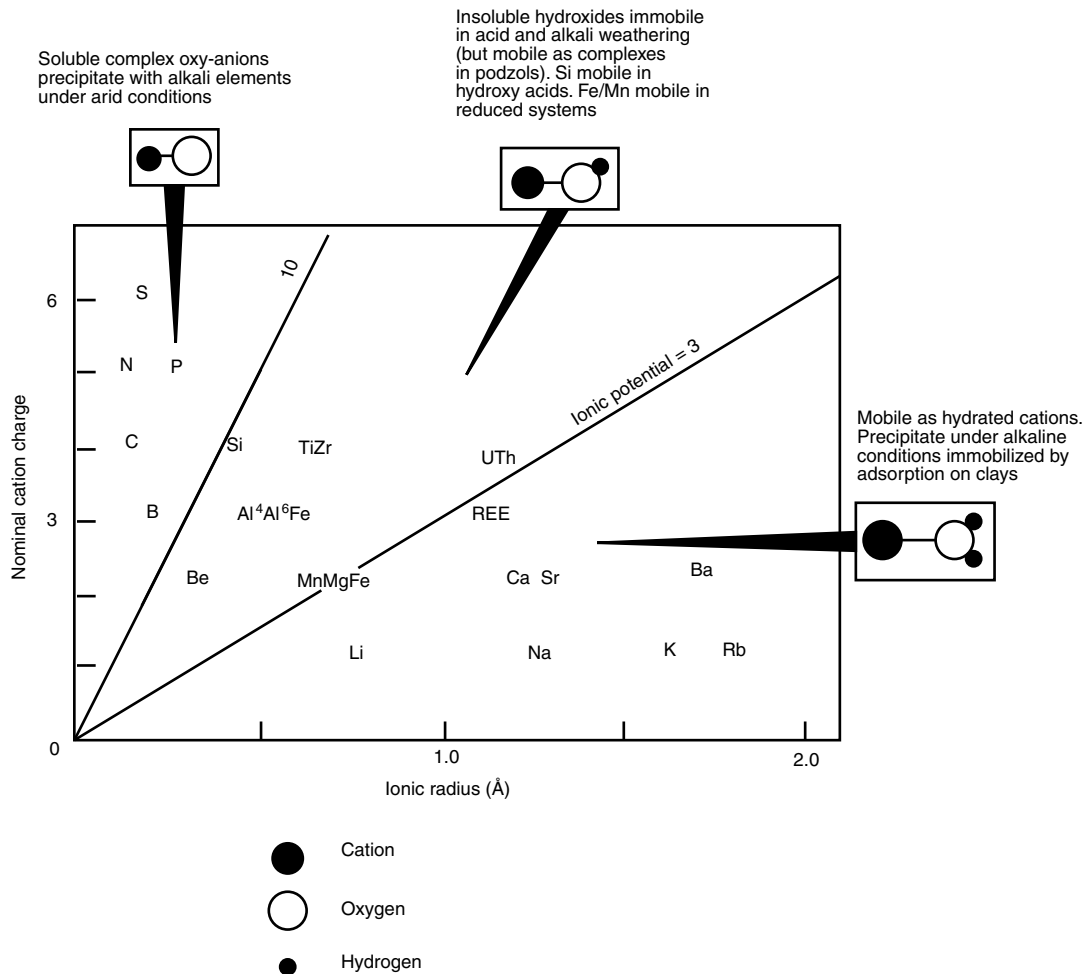


Fig. 1.4 The relationship between ionic charge and radius for various ions of importance in surface hydrogeochemical and weathering reactions. (After Gill, 1989).

weathering will leach Si from the soil profile, leaving behind a mixture of Al and Fe oxides and clays. These are *lateritic* (aka *ferralitic*) soils, common in well-drained tropical sites. By way of contrast, in highly acid soils Al and Fe are both leached to give a *podzol* with a characteristic light-coloured, silica-rich upper zone. Soils are discussed further in section 6.5.

Quartz, muscovite and K-feldspar dominate amongst clastic mineral components that survive chemical weathering from the weathered *regoliths* of igneous and metamorphic terrains (Fig. 1.5). However, the abundance of these and other primary minerals is highly variable, depending upon a number of factors including source abundance, climate and type of weathering, original grain size, rapidity of sedimentation, and so on. As usual, sedimentology cannot be reduced to mere chemistry or physics. There are also important newly formed minerals produced by weathering.

Quartz

Quartz makes up ~20% by volume of the exposed continental crust. Its crystals comprise spiral networks of linked silicon–oxygen tetrahedra, making the lattice extremely resistant to chemical attack by aqueous solutions over the acidic and neutral pH range.

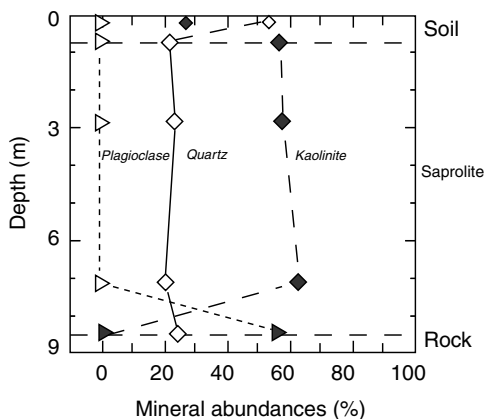


Fig. 1.5 Selected mineral abundance versus depth for the Rio Blanco Diorite, Costa Rica to illustrate the rapid changes observed in mineralogy at boundaries between bedrock/saprolite and saprolite/soil. (After Riebe *et al.*, 2003.) Note dissolution of plagioclase and neoformation of kaolinite in the regolith.

Solubility increases at $\text{pH} > 9$ when crystal surfaces are subject to increasingly effective *hydroxylation*, particularly in the presence of alkali cations from NaCl and KCl. Hydroxylation is a process that introduces one or more hydroxyl groups (OH^-) into a compound or radical, thereby oxidizing it. Polymerization occurs in supersaturated solutions to form silica gels or sols. There is much evidence that quartz dissolution is greatly facilitated (perhaps by several orders of magnitude) by dissolved Al in the porewater environment. The main route by which quartz is made dissolvable, and also fragmentable, appears to be along microcracks and fractures.

Feldspar

As noted above, feldspar is the most abundant mineral in the Earth's crust and is most notable for being a major player in soil acidification and terrestrial inorganic CO_2 uptake (Cookie 5). It is an aluminosilicate in which Si–O and Al–O tetrahedra link to form an ‘infinite’ three-dimensional framework with variable proportions of the alkali cations Na, K and Ca in the interstices. The rate of dissolution is strongly temperature dependent and is also an interesting V-shaped function of pH (Fig. 1.6). At low pH the feldspar weathering reaction is simply a transformation by acid hydrolysis of the Al bonds. The main source of protons is dissolved CO_2 , both from the atmosphere and the soil as a product of respiration. Such *acid dissolution* is probably the commonest form of soil weathering condition; it results in the precipitation of the white clay mineral, *kaolinite* and the liberation of the alkali and alkaline-earth elements in solution as hydrated ions, carbonate or bicarbonate ions with silica sometimes as byproduct (Cookie 4, Equation 4.1). Careful experiments show that the silica probably originates under slow-leach acidic conditions as a gel. Reaction rates are low at or about neutral pH, rising again because of hydroxylation at high pH when Si bonds are susceptible to dissolution in the presence of base cations. K-feldspar is also unstable under acid conditions and yields the products potassium carbonate, kaolinite and silica. There are several other possible products of the feldspar-weathering reaction depending upon local conditions of pH, which control the nature of the dissolved Al species—the most notable is *gibbsite*, $\text{Al}(\text{OH})_3$. At very low pH (< 3) reactions cause metallic cation leaching and the formation of a

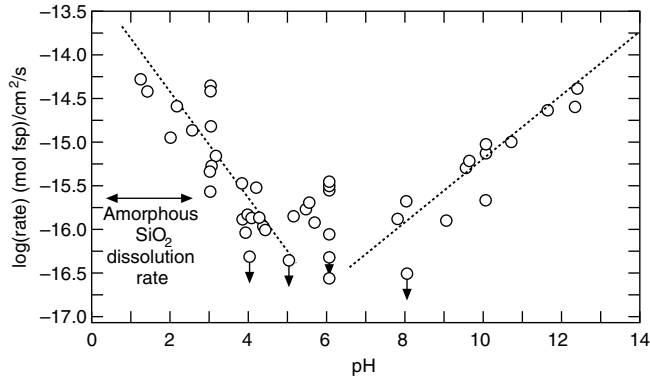


Fig. 1.6 Experimental sodium-rich feldspar (albite) dissolution rate as a function of weathering solution pH. Note the V-shaped curve common to many silicates. (After Blum & Stollings, 1995.)

thick surface layer rich in Si. Such surface layers do not form at other pH values and the dissolution process is dominated not by diffusion through a surface layer but by direct bond breaking and scavenging of ‘danglers’ (broken bonds).

Although there are a wide range of apparent activation energies and rate constants recorded in experiments, the generally observed field-weathering order for the various feldspar types is:

anorthite(Ca-plagioclase) > albite(Na-plagioclase)
> orthoclase/microcline/sanidine(K-feldspars)

There is some evidence that the dissolution of K-feldspar is inhibited by the presence of Al in solution. Figure 1.7 shows spectacular etch pits and channels formed by Holocene weathering of a K-rich feldspar.

Want to know more about chemical weathering as a geosink for global atmospheric CO₂? Turn to Cookie 5.

Micas

Micas have a distinctive sheet structure in which Mg, Al and Fe cations in octahedral arrangement lie between layers of (Si, Al)O₅ tetrahedral sheets. Cation substitutions give rise to negatively charged layers which are neutralized by an interlayer of K in both muscovite (K-rich mica) and biotite (Fe-rich mica) mica. This interlayer is released relatively quickly during weathering. Weathering reactions of the

framework ions are slower and chiefly controlled by hydration and hydroxylation at broken metal–oxygen bonds, the process occurring progressively inwards from edge faces. Muscovite exhibits a typical V-shaped dissolution rate curve with respect to pH, with the lowest rates around pH 6. Biotite shows the same dissolution rate trend as muscovite at acid pH, but rates are usually very much higher (five times that of muscovite and up to eight times that of plagioclase) because of the dissolution effects of aqueous oxidation upon ferrous iron, Fe²⁺.

Chain silicates

These comprise the pyroxene and amphibole groups in which silica tetrahedra are linked in either single (pyroxene) or double (amphibole) staggered chains by oxygen sharing. The chains are linked and strongly held together by metallic cations like Ca, Mg and Fe. Amphiboles have additional OH⁻ in the rings between opposite chains. Weathering is chiefly by surface reactions that remove the metallic cations and acid hydrolysis by protonation to form surface species bonded as silicon oxide surface species. Rates of dissolution vary inversely with pH, with actual rates varying widely in these often chemically complex groups. As in biotite mica the occurrence of transition-group metals like Fe and Mn means that redox reactions with dissolved oxygen also occur, leading to generally faster rates of overall dissolution. Oxidation of liberated Fe²⁺ leads to surface crusting of

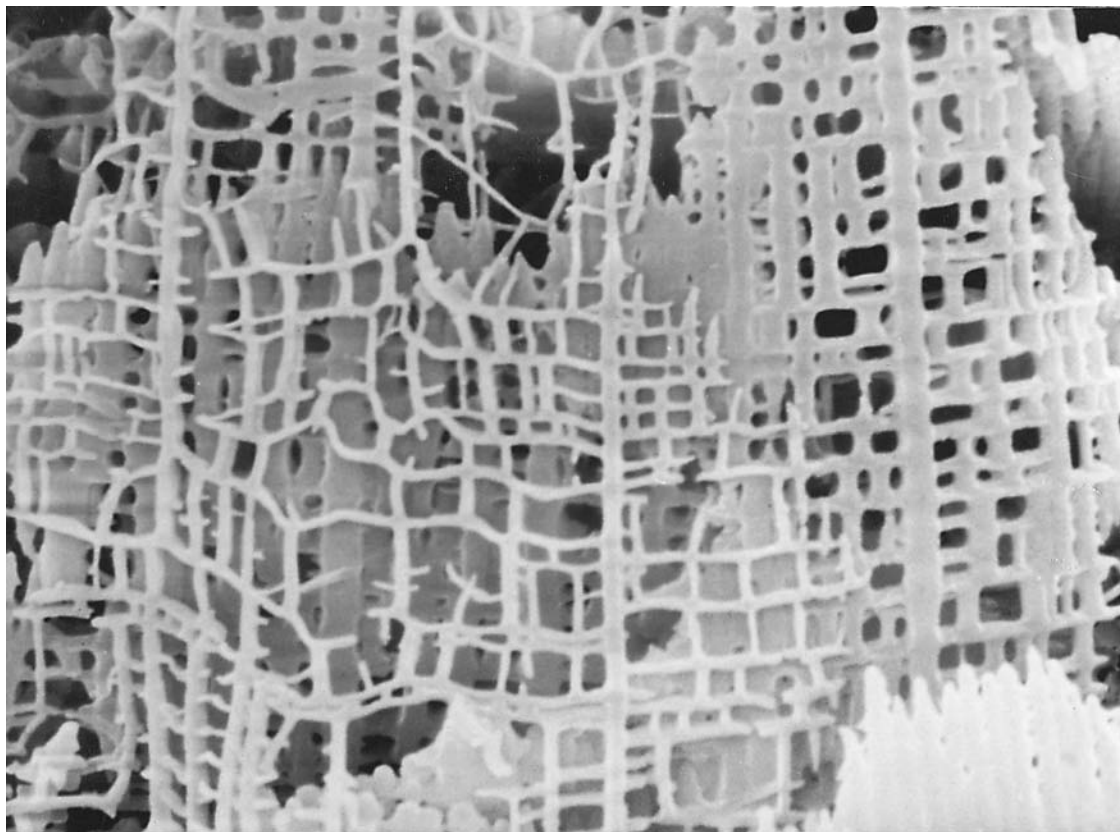


Fig. 1.7 Scanning electron microscope image of a resin cast of weathered feldspar. The former grain surface of the K-rich alkali feldspar is below the plane of the image and the weathered interior is in focus. The resin network has penetrated into the interior of the crystal along etch pits. Field of view 24 μm . (After Lee & Parsons, 1995, 1998; image courtesy of authors)

amorphous Fe^3 precipitates, but it is not thought that these adhere significantly enough to lead to diffusion-controlled reactions.

Olivine

Olivine has a simple structural arrangement of isolated SiO_4 tetrahedra linked by the divalent cations Mg^{2+} and Fe^{2+} . It comprises the end-members fayalite (Fe-rich) and forsterite (Mg-rich), with a complete solid solution series in between. Mg is removed by surface leaching and complex formation after protonation. Fe-rich olivines are highly susceptible to oxidative weathering and the formation of hydrated clay minerals. Ferric crusts of *goethite* characterize humid saturated conditions whilst *haematite* occurs under subhumid to arid

unsaturated conditions. A major role for microbial oxidation of Fe^{2+} during acid-weathering is suggested by experiments that record lowering of mineral reaction due to surface adsorption of Fe^3 .

Clay minerals

Clay minerals are the most important newly formed mineral group in the weathering zone since their eventual erosion and deposition produces copious mud-grade sediments that give valuable information about weathering conditions. We have already encountered *kaolinite*, formed under humid, acid weathering conditions from the alteration of feldspar-rich rocks. *Illite*, a potassium aluminium hydrated silicate, is formed by weathering of feldspars and micas under

alkaline weathering conditions where significant leaching of mobile cations such as potassium does not occur. *Smectites* are complex expandable sheet silicates with intracrystalline layers of water and exchangeable cations. They form from the weathering of igneous rocks under alkaline conditions. Gibbsite is simply aluminium hydroxide and forms under intense tropical weathering conditions with high annual precipitation (>2000 mm) when all other cations and silica present in bedrock are leached out.

Apatite and the P cycle

Apatite is either a stoichiometric, fluorinated calcium phosphate (igneous fluorapatite, FAP) or a non-stoichiometric sedimentary carbonate fluorapatite (CFA) with variable substitution of Mg^{2+} , Na^+ and CO_3^{2-} . The mineral is a common accessory mineral in granitic rocks and forms the majority of sedimentary phosphate rock. Both FAP and CFA define the primary exogenic phosphorus sink and serve as the long-term weathering source of P to the biogeosphere where it is an essential nutrient element for cellular life and acts as a control upon net ecosystem production. Weathering is optimal under warm, acidic pH conditions, with FAP being the easiest to dissolve.

Rock weathering profiles: changes, age and depth

Data from studies of chemical, physical and mineralogical changes with depth in weathered granodiorite are shown in Figs. 1.8 & 1.9. The fresh granodiorite comprises plagioclase feldspar (32%), quartz (28%), K-feldspar (21%), biotite (13%) and muscovite (7%) with rarer (<2%) amphibole. The main secondary minerals formed during weathering are kaolinite with lesser amounts of halloysite, goethite, haematite, gibbsite and amorphous Fe-hydroxides. Note the rapid loss of alkaline earth elements Na and Ca from fresh to plagioclase-weathered bedrock with K, Fe and Al showing slight relative enrichment. K subsequently declines in saprolite as K-feldspar is incongruently dissolved whilst Fe and Al enrich, the former due to Fe^{3+} precipitation in hydroxides, the latter due to kaolinite precipitation. Si usually behaves conservatively or slightly depletes throughout, apart from in the soil zone where it concentrates and Al/Fe strongly depletes. Cosmogenic isotope dating reveals mean rates of descent of the weathering fronts at speeds of

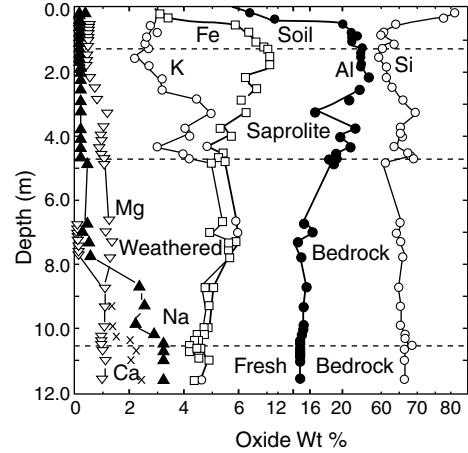


Fig. 1.8 Elemental distributions with depth in the Panola regolith developed over $\sim 10^5$ yr on a granodiorite bedrock, Georgia Piedmont province, USA. Weathering was essentially isovolumetric with preservation of primary granitic textures in the saprolite, average saprolite porosities of $\sim 35\%$, presence of boxwork textures of secondary minerals surrounding primary remnants and etch-pit formation. (After White *et al.*, 2001.)

$\sim 4\text{--}7\text{ m}/10^6$ yr. These rates are very much slower than experimental feldspar weathering rates and suggest that in old regoliths surface mineral reactivity decreases significantly with time.

Chemical index of alteration

Although chemical weathering is complex there exists a simple Chemical Index of Alteration (CIA) that is useful to assess its extent. It involves simplifying weathering of the upper crust to a combination of feldspar and volcanic glass, involving only the commonest oxides in the silicate fraction, Al_2O_3 , CaO , Na_2O and K_2O . The CIA is given by $100 \cdot Al_2O_3 / (Al_2O_3 + CaO + Na_2O + K_2O)$ and varies between 100 and 47. The pristine upper crust has a mean CIA of 47, with 100 signifying wholesale removal of all alkaline earths. CIA values calculated for the suspended load of the major rivers of the world define a chemical weathering path (Fig. 1.10; section 23.3). Rivers draining tectonically active catchments have low CIA values since physical weathering and high mechanical erosion rates predominate here.

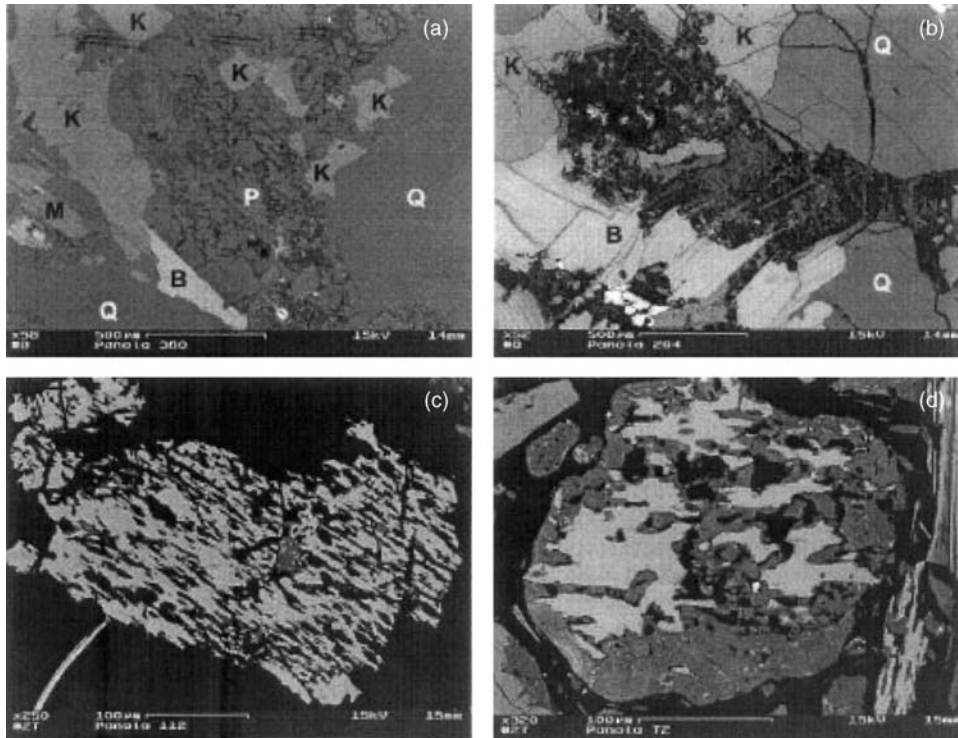


Fig. 1.9 Scanning electron microscope backscatter-mode images of rock sample thin-sections from various depths in the Panola regolith (P, plagioclase feldspar; K, –potassium feldspar; B, biotite mica; M, muscovite mica; Q, quartz). (a) 9.1 m depth; initial incongruent weathering of plagioclase feldspar in bedrock. Dark areas within the crystal are the clay mineral kaolinite. Note the pristine K-feldspar. (b) 7.5 m depth; the plagioclase crystal (centre) has virtually gone, leaving clay-filled vugs (black areas). The adjacent biotite mica is starting to weather along cleavage planes (dark areas within the biotite). The K-feldspar is again pristine. (c) 2.8 m depth. In this saprolite zone the K-feldspar has dissolved, gaining a skeletal appearance with open vugs (black areas) that originally may have been partially filled with clay. (d) 1.8 m depth just below the soil-saprolite interface. The K-feldspar is partially replaced by clay. Note the relatively unreacted muscovite mica to the right of the image. (After White *et al.*, 2001.)

Al-release from silicates by acid hydrolysis ('acid rain' problem)

Acid hydrolysis reactions in soil zones with base cations (especially Ca^{2+}) usually lead to effective neutralization of rainwater, which in unpolluted areas has a pH of around 5.0. However, in many temperate zones with high rainfall and thin soils on Ca-poor substrates, the acidity is not neutralized, leading to formation of characteristic podzol soils (see section 6.6) with leached Al-poor surface horizons. Vastly increased industrial pollutants and emissions (CO_2 , SO_2 , NO) have accelerated these processes in many areas, including exports of gases to 'innocent' countries (e.g. Scandinavia), leading

to extensive Al-release in acidic waters and widespread environmental damage. The very steep increase of Al solubility in waters of low pH is illustrated by the data of Fig. 1.11. Assessments of acidification depend heavily upon estimations of cation fluxes, sources and sinks through the water-soil-bedrock system.

1.3 Solute flux: rates and mechanisms of silicate chemical weathering

In these times of rapid environmental change it is essential to truly understand and to be able to predict the rates and mechanisms of chemical weathering of

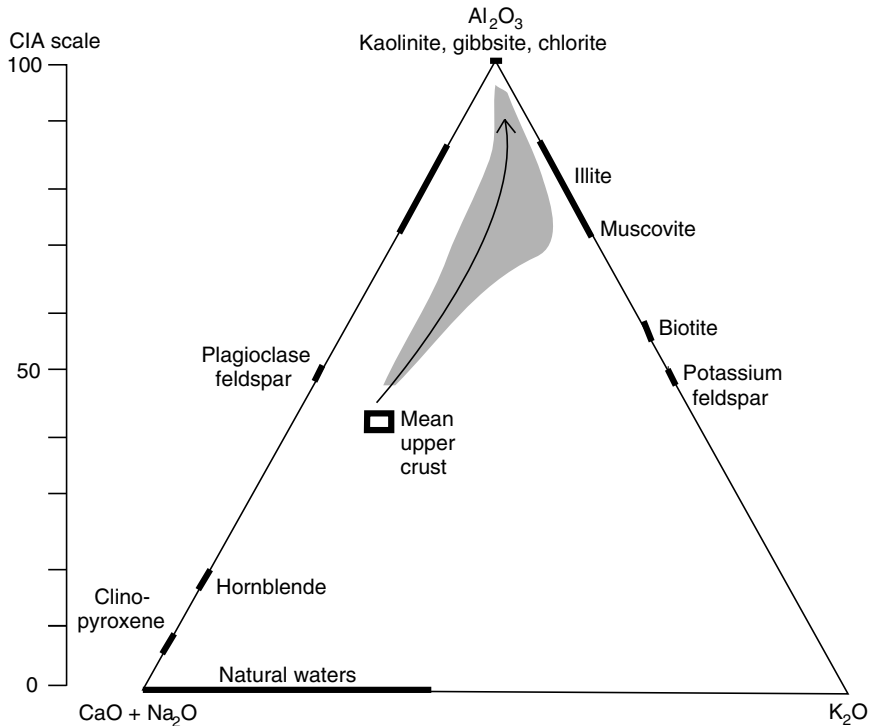


Fig. 1.10 Triangular diagram Al_2O_3 – $(CaO + Na_2O)$ – K_2O , with approximate compositional ranges of natural waters, common silicate minerals and Chemical Index of Alteration scale. (After McClelland, 1993.)

silicate-rich bedrock in response to water through-flow. Very generally the *solute flux* approach to catchment weathering seeks to draw up a balance sheet of all inputs and outputs to the catchment chemical system—it is basically a *conservation of mass* exercise. Its simplest expression states that:

$$\text{cation weathering rate} = \text{leaching} + \text{uptake} - \text{deposition} - \text{base saturation decrease}$$

It is usual to define weathering reactions in terms of a *rate constant*, the amount of mass lost from unit surface of a mineral over unit time. It is defined by *Arrhenius's law* (Cookie 6), controlled both by the energy needed to make a reaction happen, the *activation energy*, and by the *temperature* of reaction conditions. The law is easiest to understand kinetically, since any reaction needs sufficiently energetic collisions between reactant ions to cause it to happen; an *energy threshold* must be surmounted. In the case of silicate mineral surfaces in contact with natural waters, the warmer the aqueous

phase the more energetic will be the attacking protons and therefore the faster the reaction will be. Not only that, but the energetics increase exponentially as temperature increases. This means that there is a considerable (25-fold) disparity between the energy available for weathering at the poles compared to the tropics.

Want to know more about the rates and mechanisms of chemical weathering? Turn to Cookie 6.

Flow rate of water through the weathering zone is important because much rainfall is recycled back to the atmosphere by evapotranspiration; the amount available for throughflow is dependent on vegetation type and density. Most silicate reactions proceed to a point where the products have concentrations as predicted by the relevant *equilibrium constant*, which for silicates are *very low* (Cookie 6). When the rates of forward and backward reactions become equal, no

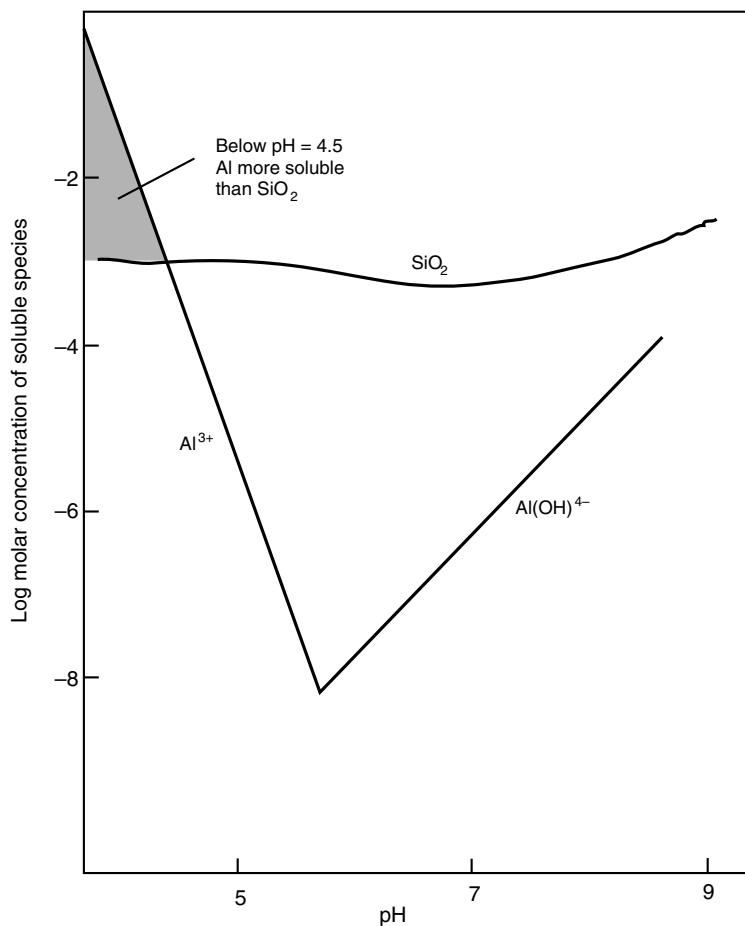


Fig. 1.11 The solubility of aluminium and silicon as a function of pH. (After Raiswell *et al.*, 1980.)

further weathering occurs. Only by removing saturated porewaters and replacing them with new water can more weathering reactions occur. Optimal conditions for chemical weathering occur in forested orogenic highland areas of the humid tropics, where very thick soil and chemically altered bedrock zones have formed in the Holocene and where soil instability due to slope failure enables periodic exposure of fresh saprolite or bedrock. Lack of physical weathering and erosion on continental plateaux lead to thick residual soils and a marked decrease in total export of ions. Such catchment slope conditions are known as *transport-limited*, for there is always an abundance of weathered product to transport.

There is much experimental work to suggest that weathering rates are linear, controlled by surface reactions between mineral and aqueous phases. Surface in this case refers to fractured and cleaved mineral faces. There is also sound evidence from electron microscopy studies that weathering can occur along uncracked and uncleaved areas of minerals, the process occurring along submicroscopic diffusion paths at sites of lattice dislocations. The chemical attack is seen in the form of etch pits (Fig. 1.7). Rates of surface reactions are usually very slow compared to diffusion rates, and the concentration of products adjacent to the reacting mineral surface are comparable to that of the weathering solution.