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Geochemistry and sedimentology of marine and nonmarine evaporites

By Hans P. Eugster[1])

ABSTRACT

Evaporite studies were initiated in the Eocene Green River formation, Wyoming and Pleistocene–Holocene Lake Magadi, Kenya. Water compositions evolve through mineral precipitation and other fractionation mechanisms and the fate of a particular brine is determined by inflow compositions, that is bedrock lithology. Computer calculations can simulate brine evolution. Deposition of chemical sediments in closed basins has been defined in terms of subfacies processes and facies complexes change in time and space. The Lake Bonneville–Great Salt Lake system serves as an example. Seawater evaporation can be monitored by computer calculations and the results agree with observations if we allow backreaction of the Ca sulfates.

ZUSAMMENFASSUNG

Ursprünglich beschränkten sich die Studien kontinentaler Evaporite auf die eozäne Green-River-Formation (Wyoming) und den rezenten Magadisee (Kenia). Sie wurden später auf viele andere Salzseen ausgedehnt, inklusive den Grossen Salzsee (Utah). Während der Verdunstung wird die chemische Zusammensetzung der Gewässer vor allem durch Mineralausscheidungen sowie auch andere Fraktionierungsmechanismen modifiziert. Die Evolution der salinaren Restlösungen hängt hauptsächlich von den Verwitterungsreaktionen im Einzugsgebiet ab. Diese Abhängigkeit kann durch Computerrechnungen dokumentiert werden. Die Ablagerung der chemischen Sedimente ist durch Prozesse bedingt, die Subfazies und Fazieskomplexe charakterisieren, welche sich in Raum und Zeit ändern. Der Verdunstungsablauf von Meerwasser kann durch Computerrechnungen simuliert werden, und die Resultate stimmen mit Feldbeobachtungen überein, falls die Kalziumsulfate mit den Restlösungen reagieren können.

My contribution to the symposium on evaporites organized by the Swiss Society of Mineralogy and Petrology on Friday, 14 October 1983 in Porrentruy consisted of a brief summary of work published during the last 25 years and of work in progress. Initial interest in the Green River formation of Wyoming, Utah and Colorado was sparked by the list of unusual authigenic minerals found to occur in the carbonate- and kerogen-rich "shales" of this formation (MILTON & EUGSTER 1959) and the fact that trona ($NaHCO_3 \cdot Na_2CO_3 \cdot 2H_2O$) is the most abundant evaporite mineral, rather than gypsum or anhydrite and halite. In order to better understand the unusual mineralogy of these continental Eocene lake deposits, we embarked on a study of active salt lakes in many parts of the world with the hope that observations of ongoing processes would

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help us interpret rocks formed in similar ancient environments. We set out to define the conditions required for the formation of alkaline brines from which trona precipitates. Lake Magadi, Kenya, became a study object, because trona has precipitated in this Rift Valley lake throughout most of the Holocene. Dilute inflow waters are of the $CaHCO_3$ type, while sulfate and chloride are comparatively low. Bedrock is fairly uniform and consists of basalt and trachyte flows formed as a consequence of rifting, with the youngest flows dated at 700,000 years b.p. By analyzing waters from the most dilute to the most concentrated stages, it was possible to show that during evaporative concentration Na and Cl are conservative, Ca + Mg and some HCO_3 are lost from solution through carbonate precipitation, most K is removed presumably through ion exchange and silica through precipitation as opaline cements (EUGSTER 1970; JONES et al. 1977). Mineral precipitation and solute acquisition through wetting–drying cycles such as those described by SMITH & DREVER (1976) are the most important factors in the chemical evolution of the brines. In desert climates, evaporative pumping (HSÜ & SIEGENTHALER 1969) of groundwaters leads to precipitation of efflorescent crusts in which all of the solutes are deposited. During subsequent wet seasons, fractional dissolution returns only the most soluble constituents to runoff and groundwater, leaving Ca–Mg carbonates and silica behind in surface and soil coatings.

To test the effect of mineral precipitation, HARDIE & EUGSTER (1970) simulated the evaporation of a wide range of dilute inflow waters with a computer program modelled after a suggestion by GARRELS & MACKENZIE (1967). The tests clearly show that the chemical evolution of a brine is predetermined by the composition of the dilute inflow, which is largely determined by bedrock lithology and weathering reactions. Modifications of the evaporation path are possible, particularly through sulfate reduction or sulfur oxidation, as shown for brines from the Chad basin (EUGSTER & MAGLIONE 1979). Computer calculations are restricted to mineral precipitation as the fractionation mechanism responsible for brine chemistry. By comparing brines from several closed basins, including Magadi, Great Salt Lake, Utah, Abert Lake, Oregon, Deep Springs Lake, Cal., Devils Lake, N.D., and Basque Lakes, B.C., EUGSTER & JONES (1979) were able to identify additional fractionation mechanisms, such as dissolution of efflorescent crusts, exchange on active surfaces, degassing and redox reactions. As summarized in EUGSTER & HARDIE (1978) and EUGSTER (1980), the great variety of brine compositions observed in salt lakes is caused largely by Ca–Mg carbonate and gypsum precipitation acting as chemical divides (DREVER 1982), with the other fractionation mechanisms acting as modifiers. Figure 1 is a summary of our current views on brine evolution.

Continental brines accumulate in the hydrologic center of closed basins which typically are located in the lee of mountain chains regardless of geographic latitude. In such settings, low valley floors provide environments of intense evaporation with some of the driest conditions associated with arctic climates, while the mountains catch precipitation, particularly in the form of snow, sufficient to recharge the system. Depending on the inflow–evaporation balance, the central basin may be occupied by a more or less perennial brackish to saline lake or by a salt pan fringed by mudflats. During the wetter stages, inflow may be largely in the form of rivers, such as at Great Salt Lake, Lake Turkana, Lake Baikal, while during the salt pan stage circulation is largely subsurface and inflow is in the form of perennial springs and ephemeral runoff. Because of
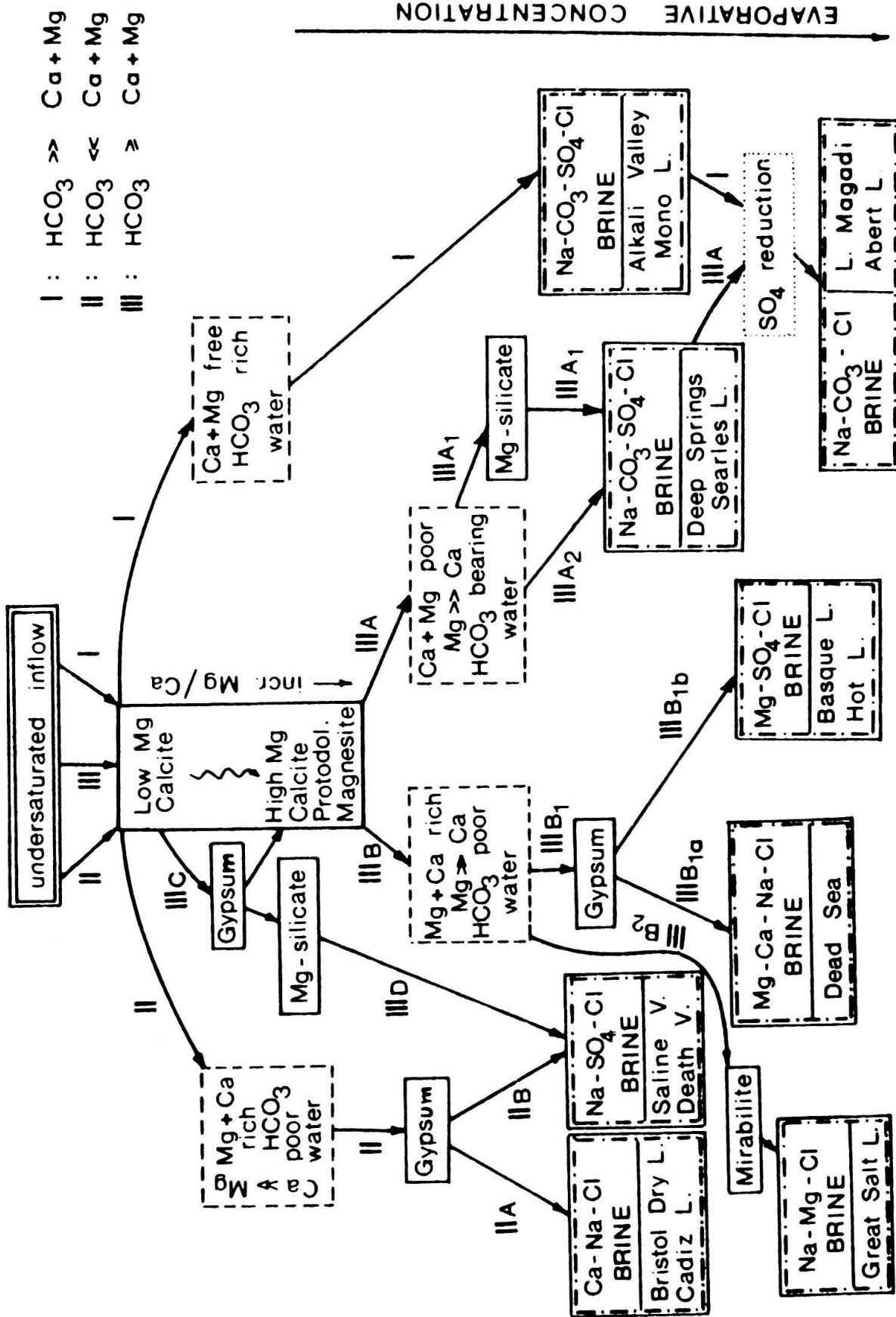
Fig. 1. Brine evolution flow diagram (from EUGSTER & HARDIE 1978), showing critical precipitates (solid rectangles) and resulting brines, together with examples of salt lakes.

the delicate response of closed-basin systems to perturbations of the hydrologic equilib-
rium, lake levels change in response to climatic and tectonic events. This is reflected in
the rapid facies changes both vertically and laterally. In fact, closed basin sediments are
dominated by transgressive–regressive cycles. To describe such systems, HARDIE et al.
(1978) have defined a number of subenvironments ranging from alluvial fans, perennial
and ephemeral stream flood plains, dunes and perennial lakes to sand flats, dry and
saline mud flats and salt pans, where each subfacies is characterized by a specific set of
sedimentary structures recognizable both in modern and ancient systems. A particular
closed basin can then be identified by an assemblage of subfacies in space and time,
forming a depositional complex. Commonly observed depositional complexes are the
alluvial fan–ephemeral saline lake complex, the ephemeral stream flood plain–dune
field–ephemeral saline lake complex and the perennial stream flood plain–perennial
lake complex. Considerable misunderstanding has arisen in the literature over our use
of the term playa-lake complex to identify the depositional environment of the Wilkins
Peak member of the Green River formation (EUGSTER & HARDIE 1975, see Fig. 2). A
playa lake (no hyphen) is a dry lake, while a playa-lake complex consists of a central
salt pan or salt lake fringed by saline and dry mud flats. The central water body
expands and retracts across the mud flats in response to changes in inflow and evapo-
ration. During the salt pan stage, brine compositions are zoned in a "bulls-eye" pat-
tern, with the highest solute loads near the center and the most dilute waters at the
fringes. A similar zonation also occurs vertically among the interstitial brines, with the
most concentrated brines near the surface where evaporation is intense. This gravita-
tionally unstable situation can persist for tens of thousands of years because the brines
are located largely in the sediment pore spaces and overturn is not feasible. In peren-
nial salt lakes, dry periods may lead to the deposition of thick salt beds, such as in the
northern arm of the Great Salt Lake at present. Subsequent flooding may lead to
stable chemical stratification with an anoxic, saline hypolimnion overlain by a more
dilute oxygenated epilimnion. Erosion of the pycnocline is very slow, especially while
saline minerals remain at the sediment surface and stratification can extend for de-
cades. Burrowers are absent, and these conditions are ideal for the accumulation of
finely laminated rhythmites, in which Ca–Mg carbonates and organic matter alternate
on a mm scale.

BRADLEY (1929) in interpreting the rhythmites of the Fossil Syncline of the Green
River formation referred to the depositional model developed for the recent Lake
Zürich sediments by NIPKOW (1923). The Lake Zürich model explains these nonglacial
varves as products of annual algal blooms, with the spring–summer blooms triggering
calcite or aragonite precipitation through CO_2 removal by photosynthesis. In the fall,
organic matter settles on the bottom, forming the dark kerogen-rich laminae of these
light–dark couplets. WEBER (1981, see EUGSTER & KELTS 1983, p. 327) has documented
the changes in water chemistry associated with such a cycle for the year 1976 in the
Greifensee, Switzerland, for waters at different levels of the water column. During algal
blooms, pH and carbonate concentration go up in the near-surface waters, leading to
20-fold supersaturation with respect to calcite. Calcite precipitation cannot keep up
with carbonate ion production until water temperatures rise to near 30 °C in the
summer months. In Lake Zürich, historical records prove that each light–dark couplet
represents one year's deposit and this fact has been used in trying to establish ages for
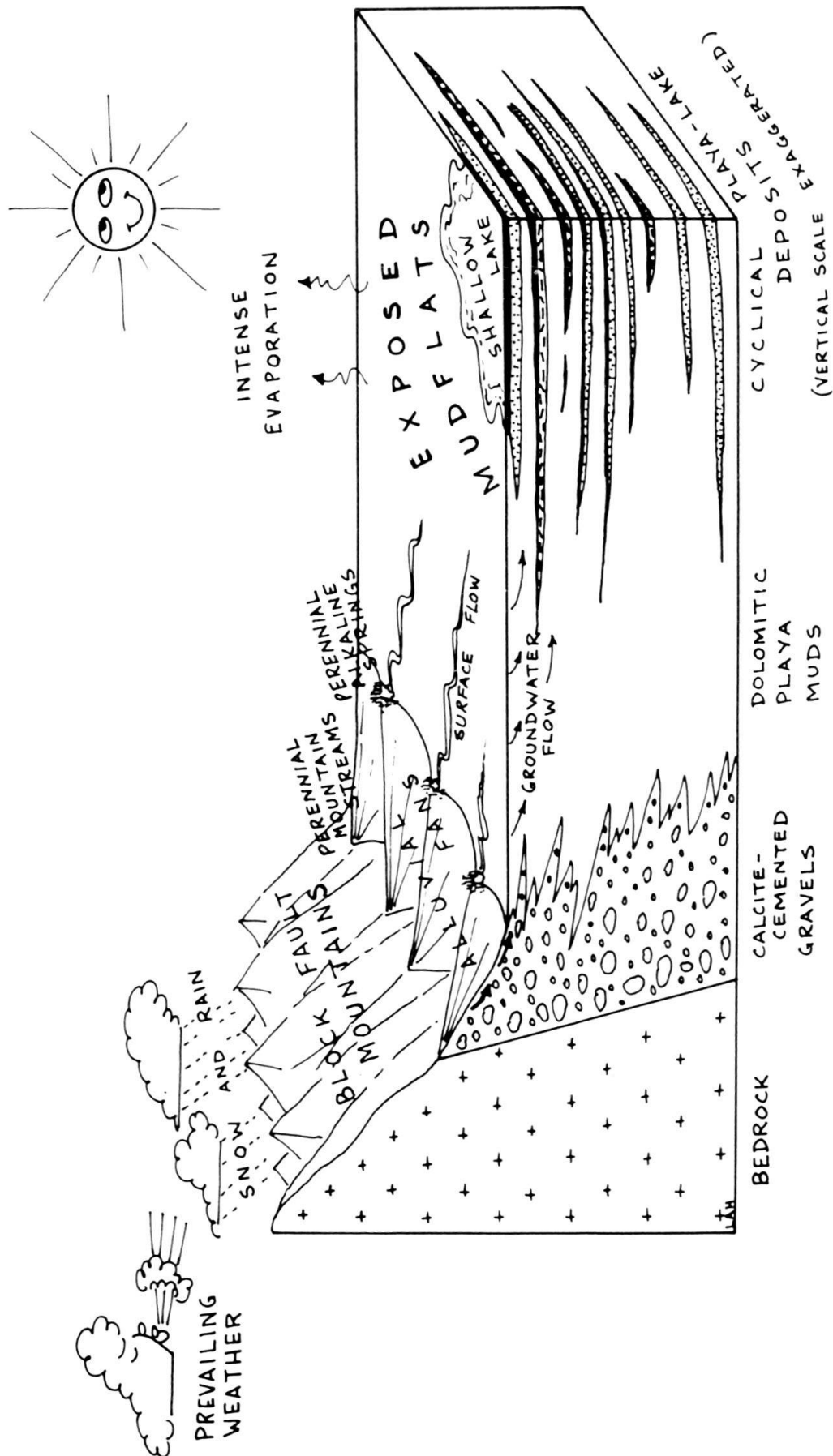
Fig. 2. Schematic block diagram of the depositional environment envisaged in the playa-lake complex of the Wilkins Peak Member, Green River Formation, Wyoming. A shallow perennial central lake is surrounded by carbonate mud flats which are normally exposed to air and are in turn fringed by alluvial fans. The lake deposits are either oil shale (black) or trona (trona stage not shown). Oil shale deposition follows either upon flat pebble conglomerates, lime sands (dots), or trona. The bulk of the deposits are carbonate muds (unshaded) (from EUGSTER & HARDIE 1975, p. 331).

similar laminites. The extrapolation is valid only if it can be demonstrated that deposition took place in a perennial water body similar to Lake Zürich. This is rarely possible. For the Wilkins Peak member of the Green River formation, EUGSTER & HARDIE (1975) showed that deposition of many laminites took place on mud flats with storm related sheetwash as transport agent. If laminations do form in a standing body of water, as probably is the case for oil shales, laminations can be destroyed by a variety of postdepositional processes, including burrowing and growth of authigenic minerals. Substantial carbonate precipitation can occur on the mud flats in the form of evaporitic crusts and soil coatings, spring and stream tufa and as cements in alluvial fans. SMOOT (1978) has documented the importance of this type of carbonate production. Pisoliths and oncoliths also are commonly found in these environments and a Recent example from Bolivia has been described by RISACHER & EUGSTER (1979). A summary of lacustrine depositional environments is given in Table 1, taken from EUGSTER & KELTS 1983.

Alkaline waters do not lead to gypsum precipitation, because calcium has been exhausted by carbonate deposition before gypsum saturation can occur. In all other waters, however, gypsum and anhydrite are the least soluble and hence the first saline

Table 1: *Depositional subenvironments of chemical and biochemical lacustrine deposits.*

A. Hydrologically open hardwater lake: fixed shoreline	
1. Littoral	Beaches, bioturbated micritic carbonate, massively bedded, with variable proportions of organic matter, siliciclastics, diatoms Charophyte chalks, ostracod, mollusc – debris and lag deposits. Bioherms, stromatolites, oncolites, cemented in part
2. Pelagic, oxygenated (full circulation)	Bioturbated micritic carbonate muds and silts with organic matter, siliciclastics and diatoms
3. Pelagic anoxic (seasonally or permanently stratified)	Carbonate (algal) laminites, interbedded with carbonate turbidites. Variable amounts of siliciclastics, diatoms
B. Hydrologically closed, perennial lake: shoreline moves	
1. Supralittoral	Laminated to thin-bedded carbonate muds, silts and sands with variable amounts of siliciclastics. Flat-parallel and lenticular laminations, scour-and-fill structures, mudcracking, burrowing, evaporative cements, efflorescent crusts, salt casts, dry mud flats.
2. Intralittoral (saline mud flats)	Transgressive–regressive sequences of carbonate muds, silts and sands. Flat-pebble conglomerates, mudcracking, bioturbation, salt disruption. Coated pebbles, pisolites, oolites and dripstones
3. Eulittoral (perennially flooded)	Laminated or bioturbated muds, silts or sands. Oncolites, bioherms, stromatolites. Carbonate–gypsum laminites, carbonate–kerogen laminites (oil shales). Bedded salts
4. Pelagic	Oxipelagic–full circulation: bioturbated muds, displaced fauna, flora, current structures, winnowing Anoxic–carbonate–kerogen laminites (oil shales), carbonate–gypsum laminites, turbidites, bedded salts
C. Ephemeral salt lake	
	As above with additional efflorescent crusts, interstitial brine precipitates, bedded salts

minerals to form. Depending on local hydrologic conditions and the nature of the sediment, they can grow in a bewildering variety of forms, from pure selenite beds representing protected lagoons to "chicken wire" nodules and desert roses formed by displacive intrasediment growth. The more soluble salts, such as halite or trona, accumulate in a standing body of brine or in a salt pan. Thick monomineralic beds can result in either setting. In salt pans, fractional dissolution and precipitation mechanisms assure separation from the less soluble constituents. Detailed discussions can be found in EUGSTER & HARDIE (1978).

Many saline minerals form as authigenic reaction products of the occluded brines with the sediment. Many of the mixed carbonates and sulfates listed in Table 2 belong to this class, including gaylussite, pirssonite, shortite, glauberite, bloedite. Their growth is caused by the expansion of brines over fresher pore fluids outward and downward in response to continued evaporation from the central pan. In these reactive environments, authigenic silicates are common products, including the sodium silicate magadiite (EUGSTER 1967) and a variety of zeolites (SHEPPARD & GUDE 1968; SURDAM & EUGSTER 1976). Magadiite is important as precurser for bedded chert (EUGSTER 1969, 1980), while the zeolites form by reaction of alkaline brines with volcanic glass. In fact, the association of magadi-type chert with lacustrine zeolites is strong indication of the presence of an alkaline, Green River-type setting. A number of such settings have been identified throughout the geologic column back to the Proterozoic, including Pleistocene Searles Lake (SMITH 1979), Triassic Lake Lockatong (VAN HOUTEN 1964), Devonian Lake Arcadie (DONOVAN 1975), Cambrian Officer Basin (PITT et al. 1980; WHITE & YOUNGS 1980) and Precambrian Damara Orogen, Southwest Africa (BEHR et al. 1983). As we predicted earlier (EUGSTER & CHOU 1973), many more will be found, particularly in the Archean and Proterozoic, where alkaline closed basins dominated by igneous and metamorphic rocks should have been common.

Most of our studies on active salt lakes have employed the simplest of tools, picks, shovels, pH meters, but for our recent work on Great Salt Lake, Utah, and its Late Quaternary history we assembled a dozen specialists to work on aspects ranging from stable isotopes and tephrochronology to pollen and ostracod records in the cores extracted from the lake. Preliminary results can be found in SPENCER (1982) and EUGSTER & KELTS (1983) and detailed documentation will appear shortly (SPENCER et al. 1984). Core sediments collected go back 30,000 years and we were able to reconstruct a lake level curve, including the prominent Bonneville, Gilbert, Stansbury and Eardley levels. Near the Pleistocene–Holocene boundary, some 12,000–15,000 years ago, a startling event took place, which is recorded in 40 cm sediment consisting primarily of carbonate, first calcite and then aragonite. Within less than 1000 years, Lake Bonneville changed from an open lake with drainage to the north at Red Rock pass to a salt lake located below the present level of Great Salt Lake, depositing thick beds of mirabilite. A climatic event or a stream capture must be responsible for this change and we are continuing our search of the cause. If the event was climatic, it would have to be observable also in other lakes of the Great Basin, such as those belonging to the Lahontan system.

Our experience with marine evaporites has been less extensive than our preoccupation with continental evaporites. Some years ago (HARDIE & EUGSTER 1971), before the discovery of the Miocene evaporites in the Mediterranean, we showed that much of the Solfifera Series of Sicily could be explained by clastic accumulation in a shallow,

Table 2: *Saline minerals of nonmarine evaporites.*

Brine type	Primary minerals		Authigenic minerals	
a)				
Na–CO₃–Cl	Halite	NaCl	Gaylussite	Na₂CO₃ · CaCO₃ · 5H₂O
	Nahcolite	NaHCO₃	Pirssonite	Na₂CO₃ · CaCO₃ · 2H₂O
	Natron	Na₂CO₃ · 10H₂O	Shortite	Na₂CO₃ · 2CaCO₃
	Thermonatrite	Na₂CO₃ · H₂O	Northupite	Na₂CO₃ · MgCO₃ · NaCl
	Trona	NaHCO₃ · Na₂CO₃ · 2H₂O	Hanksite	9Na₂SO₄ · 2Na₂CO₃ · KCl
			Aphthitalite	K₃Na(SO₄)₂
b)			Tychite	Na₂CO₃ · MgCO₃ · Na₂SO₄
Na–CO₃–SO₄–Cl	Burkeite	Na₂CO₃ · 2Na₂SO₄	Dawsonite	NaAlCO₃(OH)₂
	Halite	NaCl		
	Mirabilite	Na₂SO₄ · 10H₂O		
	Naholite	NaHCO₃		
	Natron	Na₂CO₃ · 10H₂O		
	Thenardite	Na₂SO₄		
	Thermonatrite	Na₂CO₃ · H₂O		
c)				
Na–SO₄–Cl	Gypsum	CaSO₄ · 2H₂O	Glauberite	CaSO₄ · Na₂SO₄
	Glauberite	CaSO₄ · Na₂SO₄		
	Halite	NaCl		
	Mirabilite	Na₂SO₄ · 10H₂O		
	Thenardite	Na₂SO₄		
d)				
Mg–Na–SO₄–Cl	Bischofite	MgCl₂ · 6H₂O	Bloedite	Na₂SO₄ · MgSO₄ · 4H₂O
	Bloedite	Na₂SO₄ · MgSO₄ · 4H₂O	Glauberite	CaSO₄ · Na₂SO₄
	Epsomite	MgSO₄ · 7H₂O		
	Glauberite	CaSO₄ · Na₂SO₄		
	Gypsum	CaSO₄ · 2H₂O		
	Halite	NaCl		
	Hexahydrite	MgSO₄ · 6H₂O		
	Kieserite	MgSO₄ · H₂O		
	Mirabilite	Na₂SO₄ · 10H₂O		
	Thenardite	Na₂SO₄		
e)				
Ca–Mg–Na–Cl	Antarcticite	CaCl₂ · 6H₂O		
	Bischofite	MgCl₂ · 6H₂O		
	Carnallite	KCl · MgCl₂ · 6H₂O		
	Halite	NaCl		
	Sylvite	KCl		
	Tachyhydrite	CaCl₂ · 2MgCl₂ · 12H₂O		

marginal marine setting. We encountered gypsum sandstones, conglomerates, stromatolites and chicken-wire nodules along with the spectacular selenite and balatino beds. More recently, LOWENSTEIN (1983) carried out a sedimentologic study of the Salado formation in Texas, a terminal marine evaporite rich in K and Mg salts. To be able to predict evaporation effects in these complex systems, we enlisted the help of J. H. Weare and C. E. Harvie to calculate brine-mineral equilibria at 25 °C. By extending an approach pioneered by PITZER (1973), HARVIE & WEARE (1980) were able to calculate
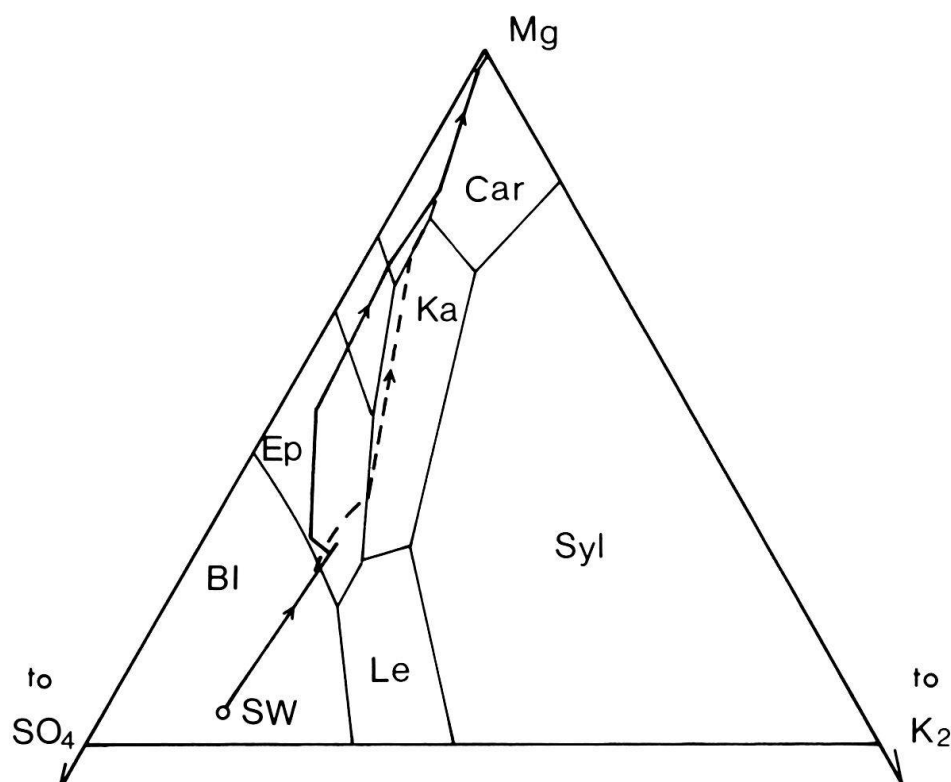
Fig. 3. Janecke diagram, $SO_4 + K_2 + Mg = 100$, halite saturated. Solid line: equilibrium path, allowing back-reaction; dashed line: fractional crystallization, no backreaction. SW: seawater, Bl: bloedite, Le: leonite, Syl: sylvite, Ep: epsomite, Ka: kainite, Car: carnallite. From EUGSTER et al. (1980).

mineral solubilities from dilute to extremely concentrated solutions regardless of the number of components present. As a test, we evaporated sea water by computer calculations (EUGSTER et al. 1980) and the results were very surprising and instructive. For equilibrium evaporation, if the calcium sulfate minerals already precipitated are allowed to back-react, glauberite and polyhalite are formed and the solution composition never enters the kainite field (see Fig. 3). The end product is the same: halite + carnallite + kieserite + bischofite, and the correspondance with mineral sequences and amounts found in natural deposits is much better than with the traditional Ca-free path based on Van't Hoff's predictions (see for instance BRAITSCH 1970). We can reproduce this latter path, which passes through the kainite field and does not involve polyhalite, simply by imposing conditions of fractional crystallization on the calculations, that is removing the calcium sulfates from interaction as soon as they precipitate. These calculations allow us to predict quantitatively what a particular mineral sequence should be for evaporation of any water in the system Na–K–Ca–Mg–SO_4–Cl–H_2O at 25 °C. Compositions of saturated solutions have been published by HARVIE et al. (1982) and recently, HARVIE (1982) has added HCO_3–CO_3 to the system. The calculated sequences can be matched against observed sequences and the discrepancies are important guides to processes other than mineral precipitation (see Fig. 4). Our studies of marine evaporites are just beginning and we hope to learn much more about these geologically important environments and deposits.
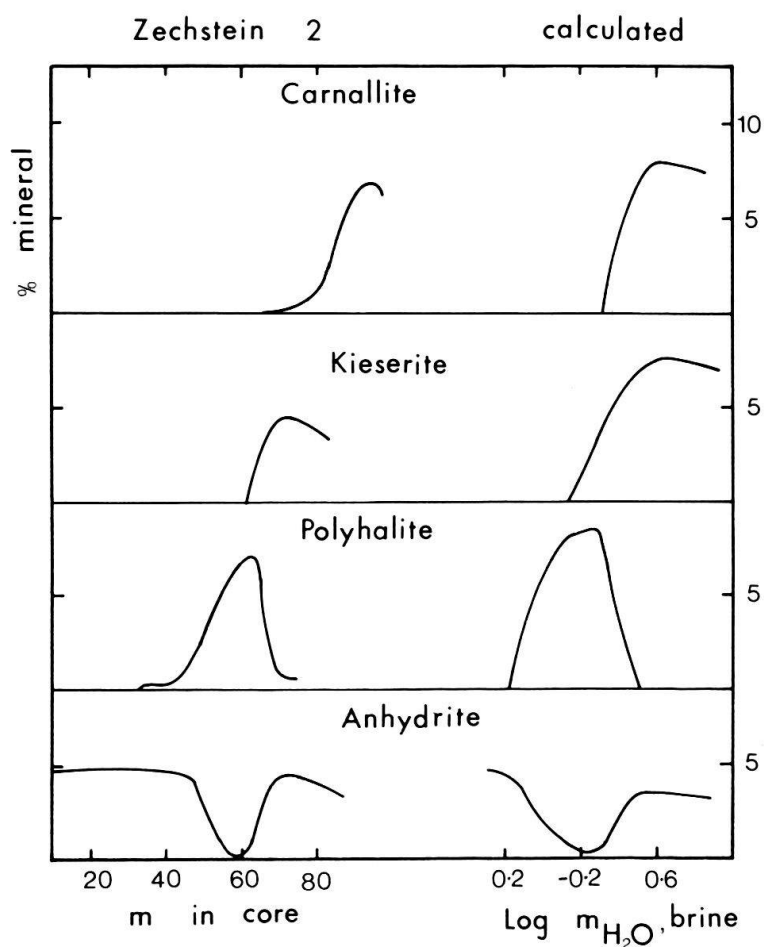
Fig. 4. Comparison of mineral abundances (with halite making up the remainder) in Zechstein II, after RIEDEL (1912), as a function of depth in the core (bottom left), with amounts expected to precipitate by calculation. Comparable stages of evaporation are indicated by depth (left) and m_{H_2O} (right).

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