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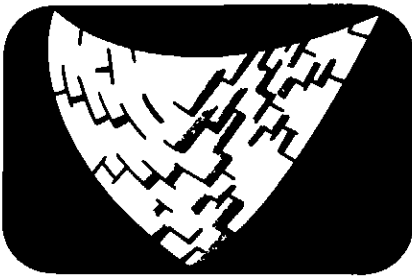
Diagenesis 2. Dolomite - Part 2 Dolomitization Models and Ancient Dolostones

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Introduction

The first paper of this review series (Morrow, 1982) was a synthesis of what is presently known concerning the chemistry of dolomitization. In that paper three parameters that are probably of major importance to the dolomitization process were identified: the Mg/Ca solution ratio; the ionic strength or salinity of the solution; and the CO_3/Ca solution ratio. Other factors, such as temperature, that influence the dolomitization process also were discussed.

In this paper I wish to focus the reader's attention on a different aspect of dolomitization - the great variety of models for dolomitization that have been proposed to explain the origin of the wide spectrum of dolostone types. In approximate historical order of appearance, these are the Hypersaline Lagoon and Reflux Model, the Burial Compaction Model, the Solution Cannibalization Model, the Coorong Model, the Sabkha Model, and the Mixed Water (Dorag) or Dilution Model (Fig. 1). The Sabkha and Coorong Models are specific to well defined environmental settings. The other models are more generalized and emphasize particular aspects of the dolomitizing process that may be operative in a variety of environmental settings. For example, in the recently proposed Dilution Model, the emphasis is on the dilution of seawater by fresh groundwater which may occur in a multitude of situations both penecontemporaneous with, or long after sedimentation.

A number of conditions must be satisfied for these models to act as agents for dolomitization (Fig. 1). These conditions, in their simplest form are as follows:

1) The amount of Mg^{2+} available for dolomitization must be adequate to form a given mass of dolostone (i.e., source of Mg^{2+}).
2) A mechanism is required to deliver the available Mg^{2+} to the dolomitization site. This mechanism may also be required to deliver a small amount of CO_3^{2-} ions to the dolomitization site and carry away Ca^{2+} ions.
3) The composition of a proposed dolomitizing solution must be conducive to dolomitization. The first paper of this review series provided some criteria to test this proposition.

Each model will be examined in the light of these conditions to place constraints on their application to the stratigraphic record and on their viability in general. A variety of specific factors may contribute to these conditions in particular instances. For example, the delivery mechanism (condition 2) may depend on porosity and permeability relationships, the hydraulic head of the solution and the time over which the mechanism is operative.

The application of these models to individual dolostone masses depends on inferences concerning the attributes of these dolostones. These include considerations of scale and spatial relationships, major stratigraphic and paleogeographic relationships, internal facies relationships, petrographic relationships, and of geochemical data. It is at this stage, where statements are made concerning the application of specific dolomitization models based on the physical and chemical attributes of dolostone bodies, that the confusion concerning the dolomitization process becomes
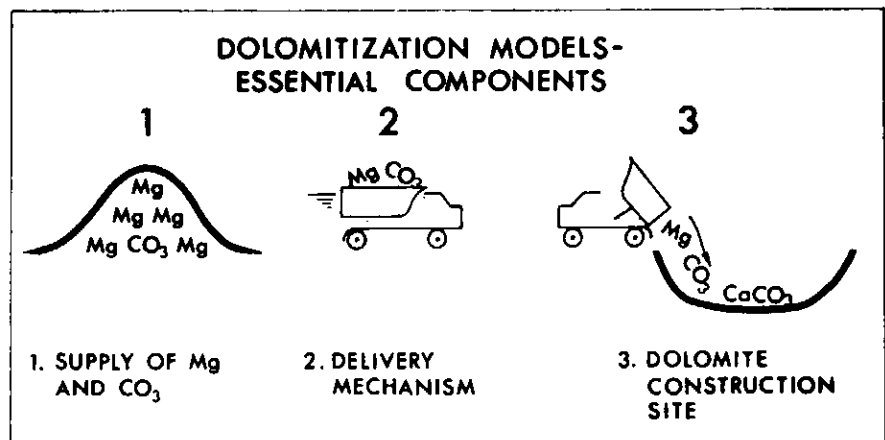

DOLOMITIZATION MODELS- ESSENTIAL COMPONENTS

1. SUPPLY OF Mg AND CO_3
2. DELIVERY MECHANISM
3. DOLOMITE CONSTRUCTION SITE

Figure 1 *A construction site analogue of the process of dolomitization illustrating the three essential elements of any dolomitization model. The external supply of magnesium, and, to a lesser extent, the supply of carbonate must be sufficient to form the observed amounts of dolomite. There must be a delivery mechanism, commonly in the form of fluid flow, adequate to deliver these ions to the dolomitization site and lastly there must exist chemical conditions favourable for the precipitation of dolomite at the 'dolomite construction site'. Failure to meet any of these conditions results in an inadequate dolomitization model.*

readily apparent to the geological public. An attempt is made here to determine the relative degree of certainty that can be placed on the diverse criteria used to substantiate the choices of dolomitization models that have been applied to many ancient dolostones in Canada (Fig. 2) and elsewhere.

Dolomitization Models

The Hypersaline Lagoon and Reflux Model. One of the first models proposed for dolomitization on a large scale was the Seepage Refluxion or more simply, the Reflux Model (King 1947; Adams and Rhodes, 1960). In this model (Fig. 3) open marine water evaporates as it passes landward across a hypersaline shelf lagoon. The resulting increase in density of these evaporated waters causes them to infiltrate the underlying sediment and to move seaward by seepage (i.e., reflux) through the seaward-dipping beds. The cycle of fresh marine replenishment of lagoon waters and seaward seepage of brines is the mechanism by which Mg^{2+} is continually transported to the sediment. The source of Mg^{2+} is the evaporated sea water itself. The dolomitization reaction probably occurs after a great deal of the gypsum (or anhydrite) has precipitated and the Mg/Ca ratio approaches 9.0 whereupon the effect of the Mg/Ca ratio overshadows the inhibiting effect of salinity (Morrow, 1978). Possibly the increase of the CO_3/Ca solution ratio that occurs during gypsum or anhydrite precipitation would also aid dolomitization.

Modern examples of the seepage refluxion hypothesis are all small in scale and operate at or near the shoreline. Müller and Teitz (1971) and Kocurko (1979) have documented shoreline dolomitization in the Canary Islands and in San Andres Island by evaporated sea water that collects in ephemeral pools in the "spray zone," a few metres above high tide. The shallow but permanent evaporating pool at Bonaire (Deffeyes *et al.*, 1965; Murray, 1969) is perhaps the best modern analogue of the large scale reflux model envisaged by Adams and Rhodes (1960).

The Mg^{2+} content of refluxing solutions in the reflux model is extremely high but it is not clear what the rate of subsurface flow of these dense solutions is likely to be. Deffeyes *et al.* (1965) calculated a rate of seaward reflux flow of 2×10^{-6} cm^3/sec. cm^2 at Bonaire but subsequent work (Murray, 1969) showed that the refluxing brines flow seaward for only a few weeks each year and most of this flow is confined to more permeable conduits bypassing most of the limestone. Model studies or computer simulations might shed light on the feasibility of large scale reflux scenarios such as that envisaged by Adams and Rhodes (1960). At present the dynamics of water movement beneath and adjacent to evaporitic shelf lagoons is unknown and a potentially fertile field of investigation. There are many examples of Canadian dolostones explained by variants of the reflux hypothesis (Fig. 2).

The Burial Compaction Model. Compaction of fine-grained sediments during burial involves the progressive explusion of pore water. Commonly large shale masses contain carbonate reef bodies and are underlain by or are bordered by thick shelf carbonate sequences. A part of this Mg^{2+}-bearing compaction water may pass through adjacent limestones and cause dolomitization (Fig. 3). The Mg^{2+} required for dolomitization is derived primarily from the pore water. Illing (1959). Jodry (1969) and Griffin (1965) were early proponents of this model (Fig. 2).

The popularity of the Burial Compaction Model waned for a long period because of the upsurge in studies of Holocene penecontemporaneous dolomites. But recent developments in the understanding of diagenetic changes in clay minerals and associated amorphous material, and the parallel maturation of organic material to hydrocarbons during burial has led to a renewed interest in this model (Davies, 1979; Mattes and Mountjoy, 1980). Clay minerals undergo a sequence of depth-related mineralogical changes that provides an additional source of pore water and of a variety of ions, including Mg^{2+}, released from clay minerals to the pore fluids. These transformations have been cited as additional causes for burial-compaction dolomitization.

There is little doubt that the kinetic inhibitions that bedevil the formation of dolomite at near-surface conditions are greatly reduced in deep burial environments largely because of the increase of temperature with depth. Also, the time

DOLOMITIZATION MODELS - CANADIAN EXAMPLES

Model	Example (Formation and author)
1. Hypersaline Basin and Seepage Reflux	Keg River - Pine Point - Presqu'ile (Skall, 1975) Muskeg - Keg River (Davies and Ludlum, 1973; Bebout and Maiklem, 1973) Prairie Evaporite - Winnipegosis (Wardlaw and Reinson, 1971) Lodgepole (Young and Greggs, 1975) Leduc (Wilson, 1975)
2. Sabkha	Charlie Lake (Roy, 1972) St. George (Collins and Smith, 1975) Turner Valley (Murray and Lucia, 1967) Muskeg-Winnipegosis (Shearman and Fuller, 1969)
3. Mixed-Water	Guelph (Sears and Lucia, 1980) Nisku (Exploration Staff, Chevron Standard Limited, 1979) Abenaki (Eluik, 1978) Allen Bay (Land *et al.*, 1975; Veizer *et al.*, 1978) Thumb Mountain (Morrow and Kerr, 1978) St. George? (Collins and Smith, 1975) Stone (Morrow, 1975; Macqueen and Thompson, 1978) Manetoe (Morrow, 1975)
4. Coorong	Belcher Group (Ricketts, in press)
5. Burial-Compaction	Presqu'ile (Griffin, 1965; Beales and Jackson, 1969) Leduc (Illing, 1959) Miette (Mattes and Mountjoy, 1980) Guelph (Jodry, 1969)
6. Solution Cannibalization	Yeoman (Kendall, 1977)
7. Tectonic or Hydrothermal	Black River - Trenton (Sanford, 1962) Keg River - Rainbow (Langton and Chin, 1968) Presqu'ile (Jones, 1980)

Figure 2 *Canadian examples of the dolomitization models discussed in this paper. Note that some units, such as the Presqu'ile Formation, have been assigned to more than one model.*

available for dolomitization is much greater in deep subsurface environments than in the near-surface environments of early dolomitization (Mattes and Mountjoy, 1980).

However, the problem of the supply of magnesium for dolomitization is an important constraint in deep burial environments. About 5.5×10^{-3} moles of Mg^{2+} are required to dolomitize 1 cm^3 of calcite at 10 per cent porosity. If this Mg^{2+} is supplied by compaction of adjacent shales and if the pore solution has the composition of seawater, then about 32 cm^3 of shale, compacted from 70 per cent to two per cent porosity, are required to provide this amount of Mg^{2+}. (These mass balance considerations were first discussed by the present author at the C.S.P.G. Andy Baillie Symposium, 1979). This assumes that all the water of compaction passes through the carbonates. In reality more than half of shale compaction water is lost vertically upward into the body of water from which the shale was deposited and most of the remainder passes downward (Magara, 1976, p. 92-95). On a basinwide scale, therefore, a more reasonable estimate would be that on the order of hundreds of cm^3 of compacted shale are required for the dolomitization of 1 cm^3 of laterally adjacent limestone. Therefore shale compaction water is insufficient to form large masses of dolomite such as the Paleozoic platform dolomite sequences of Western Canada and elsewhere.

In environments of burial greater than 2000m the transformation of montmorillonitic clays to illite is accompanied by the release of Si^{4+}, Fe^{2+}, Ca^{2+}, and Mg^{2+} ions to the pore solution so that authigenic ankerite, ferroan dolomite and chlorite are precipitated as cements in some sand-shale sequences (Boles, 1981). It seems very unlikely, however, that there is a sufficient volume of fluid at these depths to move these ions laterally and to participate in the solution of large amounts of limestone and contemporaneous precipitation of dolomite. Shale porosity is less than 20 per cent at burial depths exceeding 2000 m and processes such as aquathermal pressuring or the release of structured water from clays do not add significantly to this volume of interstitial pore water (Magara, 1976). The existing evidence strongly indicates that the late diagenetic release of ions from clay minerals is accompanied by local cementation rather than the long distance transportation of ions to shelf carbonates facing shale basins.

Another factor limiting the dolomitizing potential of these deep burial diagenetic solutions is the fact that Ca^{2+} ions are released to solution in amounts nearly
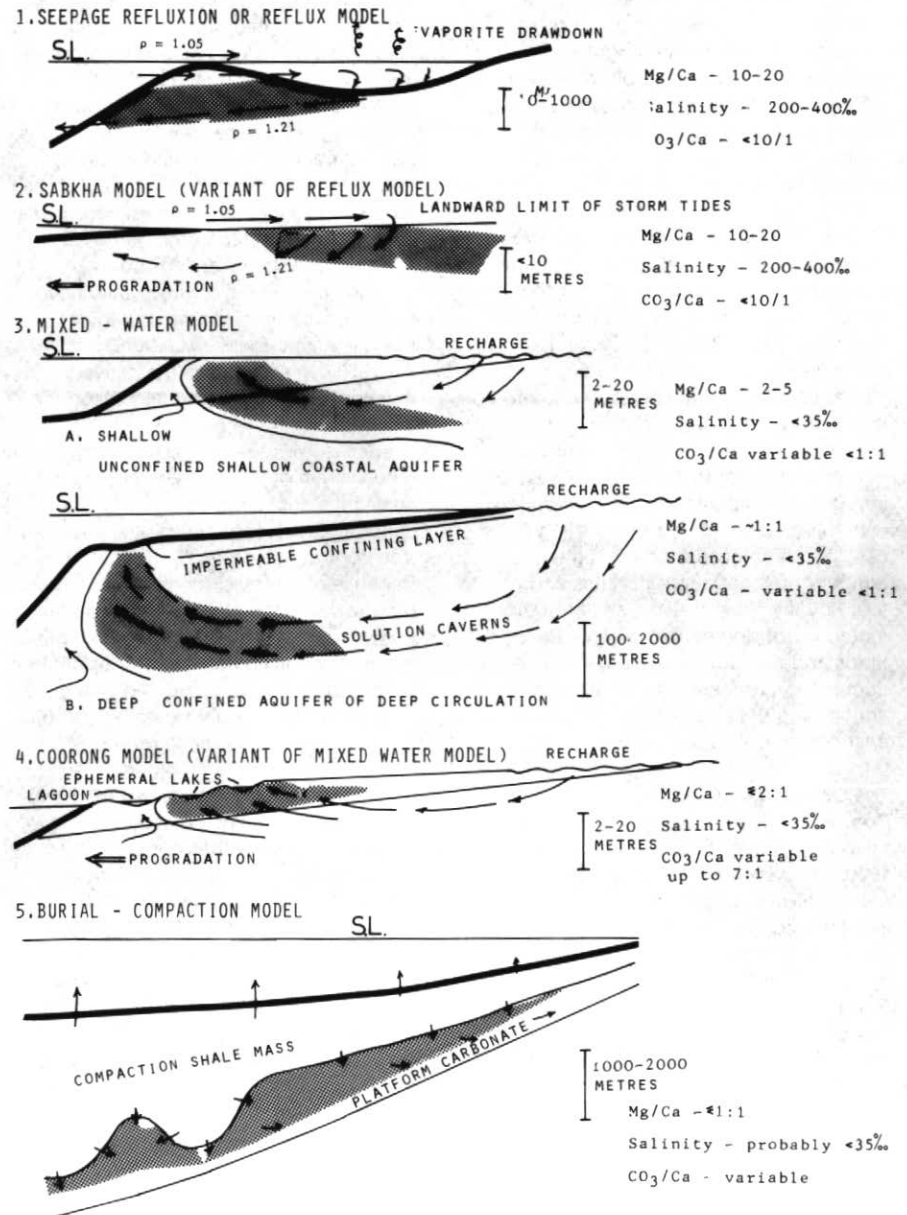
DOLOMITIZATION MODELS

Figure 3 *Major dolomitization models summarized in generalized representative cross sections. The patterned areas represent zones of active dolomitization during the operation of these models. The horizontal dimensions of these cross sections is proportional to their vertical dimensions so that systems 10s of metres thick may extend several kilometres whereas systems 1000s of metres deep may extend 100s of kilometres laterally. Arrows correspond to directions of fluid movement.*

equal to, or greater than, Mg^{2+} (Boles, 1981; Foscolos and Kodama, 1974). There is little, if any, excess Mg^{2+} available for dolomitization. In the most favourable situation one might expect the void spaces in porous carbonates facing shales (e.g., Wong and Oldershaw, 1981) to be partly occluded with ferroan dolomite, but little actual dolomitization of pre-existing limestone.

Although more work needs to be done to quantify the many variables involved in the Burial-Compaction Model, as a first approximation it does not appear that regional bodies of dolomite can form by this mechanism. The observation that many deeper water slope and basinal carbonates commonly are not dolomitized even where they are overlain by thick shale masses tends to confirm this impression (Wilson, 1975). Small amounts of dolomite may be associated with this model but in general the greater ease of dolomitization at the higher temperatures of deeper burial is countered by the progressive loss of the raw materials for dolomitization (i.e., the pore fluids and their ions) as burial-compaction proceeds. Little is known concerning the chemistry of solutions in environments of deep burial.

The Coorong and Sabkha Models. The Coorong Lagoon was known to be an area of dolomite formation since the later 1920s (Mawson, 1929) but was ignored until Alderman and Skinner (1957) inaugurated a series of investigations. Recent work (Von der Borch, 1976; Von der Borch and Lock, 1979; Von der Borch, et al., 1975 and Muir et al., 1980) has established the Coorong Lagoon as a model for early dolomitization in many ancient sequences of aphanitic dolomite that are not associated with evaporites (Fig. 3). The lagoon and a series of ephemeral lakes extend 200 km parallel to the south coast of Australia and are confined between Pleistocene fossil dunes that extend parallel to the coast. Surface drainage follows the existing topography, but the subsurface groundwater flows seaward perpendicular to the surface drainage (Fig. 3). The ephemeral lakes are filled during the humid winter months by groundwater seepage and are evaporated to partial or complete dryness during the summer months.

Extremely fine-grained calcian dolomite (yoghurt) mud and magnesite forms in the landward ephemeral lakes under the influence of continental groundwater or in the zone of mixing between sea water and continental water. This dolomite is in the form of 0.5 to 1 μm, amorphous, spherular aggregates and may have precipitated directly as a gel (Von der Borch

and Jones, 1976). Aragonite and calcite predominates in the Coorong Lagoon itself which is filled with sea water. Dolomite (and magnesite) precipitation in the ephemeral lakes is aided by ground-water dilution and by the high CO_3^{2-} concentration of the alkaline groundwater (Morrow, 1982). The source of Mg^{2+} ions has not been identified with certainty; dolomites forming close to the coast may have derived their Mg^{2+} directly from sea water but dolomites forming further inland probably utilize Mg^{2+} in the groundwater, some of which may have been derived from the weathering of basic volcanic rocks (Von der Borch, et al., 1975). In either case the delivery mechanism is the continual subsurface groundwater flow.

Successive regressions of a Coorong-type system of lagoons and ephemeral lakes on a subsiding continental margin can build thick sequences of evaporite-free peritidal dolomite (Muir et al., 1980). Because of the predominant role of fresh groundwater, the Coorong Lagoon Model may be regarded as a specific type of the more generalized mixed-water or dilution model. Only one documented Canadian example of this model exists (Fig. 2) but more undoubtedly will be found.

The other well-documented setting for penecontemporaneous dolomite, the sabkha environment, is restricted to those dolomites with accompanying evaporite minerals. Illing et al. (1965) in collaboration with geologists of the Shell Oil Company provided the first comprehensive report on the Holocene intertidal and supratidal dolomite that had replaced aragonitic sediments exposed on sabkha surfaces bordering the Qatar Peninsula on the Persian Gulf. This was followed by the studies of Kinsman (1966), Butler (1969), de Groot (1973), Bush (1973), Hsü and Schneider (1973), Patterson (1972) and Patterson and Kinsman (1981, 1982) of the Abu Dhabi Sabkhas east of the Qatar Peninsula.

Dolomite in the sabkhas of the Persian Gulf is most abundant as 1 to 5 μm euhedral crystals in the high intertidal zone close to the strandline and is strongly controlled by the network of flood channels across the sabkhas. Storm driven flood tides reach furthest inland along these channel courses (Patterson, 1972). The frequency of flooding decreases landward across the sabkha but the Mg/Ca ratio of the floodwaters rises uniformly landward by means of gypsum precipitation so that the zone of optimum dolomite formation is less than a kilometre wide straddling the boundary between high intertidal and supratidal areas (Patterson, 1972; Bush, 1973; and Fig. 3). The dense Mg^{2+}-bearing hypersa-

line floodwater brines sink downward and flow seaward through the sediment by seepage refluxion. Dolomitization of the underlying intertidal and subtidal sediments occurs to a depth of 2 to 3 m beneath the sabkha surface in regions landward of the continuous algal mat (Patterson, 1972; Bush, 1973; de Groot, 1973). Hsü and Siegenthalar (1969), Hsü and Schneider (1973) and McKenzie et al. (1980) have developed an alternative model for sabkha dolomitization based on a process which they term *evaporative pumping*. In this process, a continual flow of seawater moves landward through the sabkha sediments to replace groundwater lost by evaporation at, or near the sabkha surface. This flow pattern is the reverse of seepage refluxion (Fig. 3) but the field evidence for evaporative pumping in the sabkhas of the Persian Gulf is contradictory (see Hsü and Schneider, 1973; McKenzie et al., 1980; Patterson, 1972; Patterson and Kinsman, 1981). It is unlikely that evaporative pumping has contributed greatly to sabkha dolomitization because evaporation of sabkha groundwater is slowed considerably by a number of factors such as absorption of water at night and high relative humidity of the overlying air mass. Consequently this mechanism does not provide the volume of flow of ion-bearing groundwater necessary for dolomitization. The Sabkha Model may be regarded as a specific example of the Reflux Model modified slightly by the process of evaporative pumping. Many Canadian examples of dolomitization by the Sabkha Model have been identified (Fig. 2).

The Mixed-Water or Dilution Model. This model, in its general form, arose in the period following 1970. Before that time there were a few reports of dolomite that formed in low salinity environments such as in lakes (e.g., Klähn, 1928) or in caves (e.g., Holland et al., 1964) but Hanshaw et al. (1971) documented the first large-scale system of deep burial dolomitization under the influence of fresh groundwater in the deep confined Tertiary carbonate aquifer of Florida (Fig. 3). They emphasized the role of active groundwater circulation in supplying marine-derived Mg^{2+} for dolomitization.

Land (1973) argued convincingly from field relationships and petrographic and chemical evidence, that the Middle Pleistocene reef rocks of Jamaica were dolomitized penecontemporaneously by the interaction of marine and fresh groundwater in a shallow coastal aquifer and suggested that many ancient platform dolomites which lack evaporites and are depleted in both trace elements (e.g.,

Na[+], Sr[2+]) and heavy isotopes (^{18}O, ^{13}C) may have formed in a similar manner. This suggestion has been followed recently by many authors (e.g., Badiozamani, 1973; Land et al., 1975; Veizer et al., 1978; Randozzo and Hickey, 1977; Dunham and Olson, 1980; Choquette and Steinen, 1980). In most of these examples thick platform dolomite sequences consist of many individual regressive carbonate sediment wedges that have been penecontemporaneously dolomitized by the shallow coastal freshwater aquifers that accompanied their seaward progradation (Fig. 3). The vast differences in the chemical and isotopic composition between dolomites that have formed under the influence of freshwater and dolomites that have formed from hypersaline brines has led to an emphasis on these types of data as criteria in choosing between the Mixed-Water Model and other models. However, uncertainties in the interpretation of chemical and isotopic data preclude their use in model discrimination in the absence of other data (Land, 1980). Modern settings where the Mixed-Water Model may be operative include the Coorong area (Morrow, 1982) and Andros Island (Gebelein et al., 1980).

In the Mixed-Water Model, dolomitization occurs within the zone of mixing of fresh groundwater with phreatic seawater. The Mg[2+] ions for dolomitization are derived primarily from seawater and the delivery mechanism is the continual circulation of seawater induced by the flow of fresh groundwater (Land, 1973). Dilution of saline solutions causes slow precipitation which favours dolomite precipitation (Folk and Land, 1975; Morrow, 1982) and the high CO_3^{2-} concentration in many dilute continental groundwaters may also promote the precipitation of dolomite (Lippman, 1973; Morrow, 1982). This model has been used to explain the origin of many ancient dolostones in Canada (Fig. 2).

Solution Cannibalization and Pressure Solution Dolomite
Solution Cannibalization (Goodell and Garman, 1969) refers to the derivation of Mg[2+] for dolomitization from the dissolution of magnesian calcite and reprecipitation of low-magnesian calcite. Some examples of partial dolomitization, such as dolomitized burrow fillings (Kendall, 1977) have been explained by solution cannibalization but for more complete dolomitization other sources of Mg[2+] are required (Hsü, 1966).

Recently the phenomenon of pressure solution along solution seams and stylolites has been cited as a major cause of dolomitization (Logan and Semeniuk, 1976; Wanless, 1979). The source of Mg[2+]

ions in pressure solution dolomitization is by solution cannibalization of Mg[2+] from pre-existing magnesian calcite during pressure solution (Wanless, 1979). Complete dolomitization is thought to result from the concentration of dolomite along solution seams as a relatively insoluble residue. Intriguing as this dolomitization model is, it suffers from the difficulty in reconciling the deep burial setting of most stylolites (e.g., Mossop, 1972) with the common pre-burial conversion of magnesian calcite to low-magnesian calcite. For large amounts of dolomite other sources of Mg[2+] are required.

Tectonic or Hydrothermal Dolomite
Many coarsely crystalline, white sparry dolostones that have cross-cutting relationships with the enclosing strata have been designated as 'tectonic' or 'hydrothermal' dolomites in older studies, particularly in instances where these dolostones occur near known faults (e.g., Sanford, 1962). However, the common absence of other hydrothermal minerals implies that many of these dolostones have other origins. The relationship of some dolostone masses to faults may merely reflect the fact that groundwater movement in aquifers tends to be much more rapid along subsurface fault zones (i.e., groundwater cascade zones), enhancing diagenesis.

Ancient Dolostones - Criteria for Recognition of Dolomitization Models
Criteria that may guide the researcher in his choice of dolomitization models to apply to ancient dolostones can be divided into three broad categories - stratigraphic and facies criteria, petrographic criteria and geochemical criteria. Few, if any, of these criteria are unequivocal in themselves and it is only after the accumulation of much complementary data that a truly definitive choice can sometimes be made.

Stratigraphic and Facies Criteria
Size and Shape. Small intraformational bodies of dolomite may have local sources of Mg[2+], derived from the sediment itself as in the Solution-Cannibalization Model (e.g., Kendall, 1977; and Gebelein and Hoffman, 1973) but large masses, such as entire formations or major parts of a formation, invariably require external sources of Mg[2+]. Dolostone masses that conform to rock stratification may indicate that dolomitization occurred before burial, but non-conformable dolostone bodies that intersect primary stratification and facies at high angles strongly indicate that dolomitization is post-lithification and post-burial, particularly where these dolostones are associated

with solution-collapse breccias (e.g., Collins and Smith, 1977; Morrow, 1975). The form and location of some dolostone masses conform to known faults indicating a late, post-burial mode of origin (e.g., Skall, 1975; Jones, 1980).

Associated Evaporites and Shales. Laterally adjacent or overlying evaporites provide sources of Mg[2+]-bearing hypersaline solutions capable of dolomitizing by seepage-refluxion particularly where shelf carbonates separate evaporite bodies from more open marine sediments (e.g., Skall, 1975; Wilson, 1975, p. 317). Dolomitized reefal masses encased in evaporites is a common situation where early diagenetic brine reflux may be suspected (e.g., Sears and Lucia, 1980).

Late diagenetic movements of Mg[2+]-bearing connate brines derived by expulsion during burial compaction from evaporite masses into laterally adjacent carbonates (Jodry, 1969), or overlying carbonates (Jones, 1980) may also cause dolomitization. Similarly shales adjacent to dolomitized carbonates are also suspect as sources of dolomitizing solutions during their compaction.

Unconformities. Dolostone bodies are commonly developed beneath regional unconformities (Sonnenfeld, 1964). Many of these dolostones are best explained by the Mixed-Water Model where the unconformity surface represents the area of subaerial recharge for a fresh water aquifer (e.g., Collins and Smith, 1975 and Fig. 3). In other instances, such unconformities are considered merely to represent a period of karst development that enhances the porosity of the underlying carbonates so that dolomitizing solutions, whatever their origin, subsequently were localized in the strata beneath the unconformity (e.g., Skall, 1975).

Facies Development. Models for penecontemporaneous dolomitization such as the Sabkha and Coorong Models are verified most readily by the identification of associated facies characteristic of these environments. The sabkha depositional model with its peritidal and supratidal vertical sequence of dolomite that culminates in an interval of displacive anhydrite, forms a very distinctive cyclic sequence of lithologies and sedimentary structures (e.g., Wood and Wolfe, 1969). Similarly, environmental conditions of the Coorong Model generates an evaporite-free, cyclic sequence of very finely crystalline, penecontemporaneous dolomite with different sedimentary structures (Muir et al., 1980). In many instances reef buildups or carbonate mud mounds are

facies that coincided with paleotopographic 'highs' and thus were susceptible to dolmitization by the Reflux Model or by the Mixed-Water Model after exposure by relative changes in sea level (e.g., Eliuk, 1978; Sears and Lucia, 1980; Exploration Staff, Chevron Exploration Staff Ltd., 1979).

Paleogeography. There is a strong correlation between paleogeography, as expressed by rock facies, and dolomitization reflecting a common tectonic control (Wilson, 1975; Dunham and Olson, 1980). Shelf areas tend to be exposed during regression and become the sites for fresh-water recharge and mixed-water dolomitization. Deeper water carbonates which are never exposed, are not subject to this type of dolomitization. Thick sequences of shelf dolomite may be built up as a consequence of multiple regressions. The seaward progradation of each regressive cycle is accompanied by seaward expansion of the coastal aquifer so that a dolomitization front also moves seaward as progradation continues (Fig. 3).

Petrographic and Textural Criteria
Crystal Size. Previous reviews (e.g., Folk, 1973) have emphasized that petrographically, dolostones are of two basic types separated by a crystal size boundary of 10 to 20 μm. Dolostones more finely crystalline than this were characterized as primary or very "early", pre-burial replacements in which sedimentary and organic fabrics are well preserved (Fig. 4), whereas more coarsely crystalline dolostones that occur mainly as strataform replacements of limestone with poorly preserved fossil remains were considered to be 'secondary', 'sucrosic' or 'late replacement' dolomites (Fig. 5). A third major petrographic variety of dolomite, which has received considerable attention in recent years, is the white, very coarsely crystalline dolomite or megacrystalline (i.e. ranging from millimetres to centimetres) that, in many places, is associated with lead-zinc mineralization (Fig. 6). Much of this type of dolomite is void-filling and displays curved crystal faces (e.g., Radke and Mathis, 1980).

Dolomicrite. The very finely crystalline fabric preservation type of dolomite in many ancient dolostones is directly analogous to the fine grained dolomitic sediments that form as early replacements of aragonite in the intertidal and supratidal areas of modern carbonate tidal flats such as in the Caribbean (dolomitic crusts), in the sabkhas of the Persian Gulf and in the Coorong region of Aus-
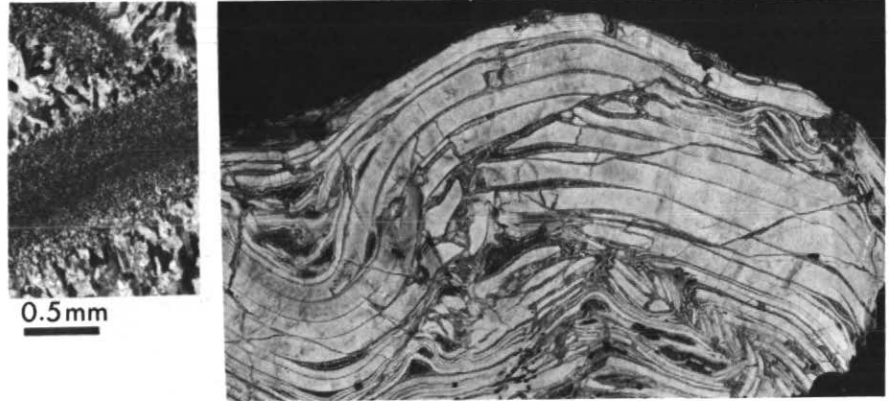

Figure 4 *A hand specimen of a bright yellow, supratidal flat dolomicrite that has undergone synsedimentary folding after partial lithification, from the Rowatt Formation, a Precambrian unit in the Belcher Islands, Hudson Bay. Thicker laminae are about one centimetre* thick. The photomicrograph on the left shows the aphanocrystalline character of this type of early replacement dolomite with preservation of microscopic detail (photographs supplied by B.D. Ricketts).
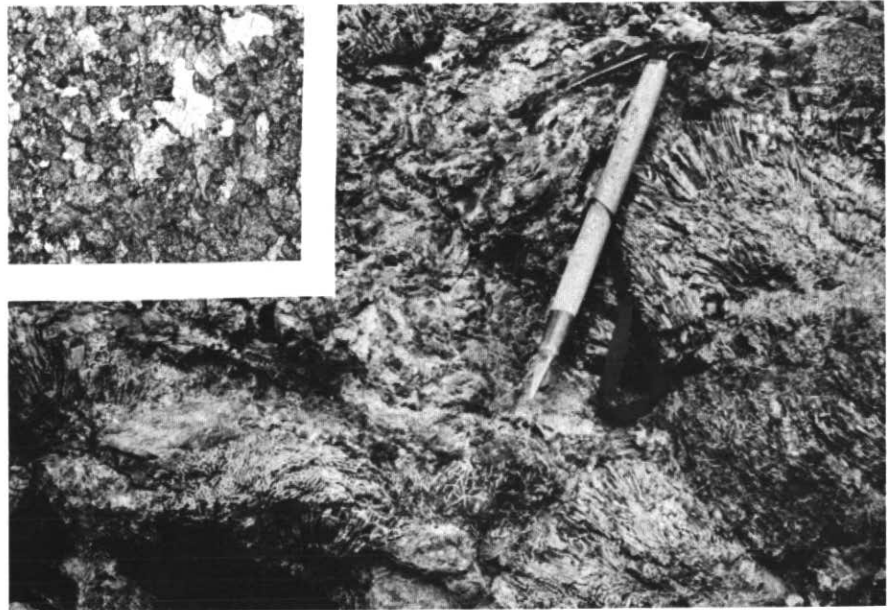

Figure 5 *Medium to coarsely crystalline 'late' diagenetic sucrosic dolomite in the Ordovician Allen Bay Formation of the Arctic Islands. Macroscopically identifiable Halysitid-type colonial corals populate this outcrop from the south coast of Devon Island. Microscopic* details of these corals have been destroyed by dolomitization that formed the hypidiotopic fabric of subhedral crystals shown in the photomicrograph in the upper left with a field of view of 1.2 mm (photographs supplied by U. Mayr).

tralia. This petrographic type of dolomite usually is inferred to have replaced aragonite mud before burial (e.g., Schmidt, 1966 and Fig. 4).

Medium Crystalline Sucrosic Dolomite. The origin of the finely to medium crystalline, sucrosic dolomites (generally hypidiotopic - an aggregation of subhedra) is much more problematic because of the apparent absence of modern analogues. However, this type of dolomite does occur as replacements of

Pliocene-Pleistocene limestones on some Caribbean Islands (Land, 1973; Supko, 1977; Sibley, in press). Unlike similar dolostones in more ancient rocks, the geological history of these Caribbean dolostones is known with a high degree of certainty and this history in conjunction with other data, suggests that most, if not all, of the Caribbean dolomite originated by the Mixed-Water Model (e.g., Land, 1973).

Crystals of this type of dolomite commonly contain cloudy centres sur-

rounded by clear rims and the Caribbean dolostones are no exception. Sibley (1980) suggested that this phenomenon, in the Caribbean examples at least, is due to the evolution of the diagenetic pore fluids from a state of near calcite saturation to a state of calcite under-saturation, as grain growth continues under the influence of mixed-water diagenesis. It is likely that the interlocking "hypidiotopic" texture of subhedra and anhedra that characterizes ancient sucrosic dolostones is the result of grain growth from an initial state where very finely crystalline dolomite euhedra are scattered throughout. These crystals grow and eventually join along irregular compromise boundaries (e.g., Morrow, 1978). The euhedral, micrometre sized dolomite rhombs that are scattered in the recent sediments forming Andros Island (Gebelein et al., 1980) may record the beginning of a progression towards a totally dolomitized fabric of sucrosic dolostone (Fig. 7).

Although many ancient, stratiform, platform dolostone sequences of sucrosic, dolostone have been interpreted to be the consequence of mixed-water diagenesis (e.g., Randazzo and Hickey, 1978; Dunham and Olson, 1980), the Reflux Model has been invoked in situations where sucrosic dolostones are adjacent to evaporites (e.g., Sears and Lucia, 1980) or adjacent to shales (Mattes and Mountjoy, 1980).

Megacrystalline White Dolomite. This type of dolomite is less abundant than the more finely crystalline varieties but is economically important as a host rock for lead-zinc deposits (e.g., Presquile Dolomite) and hydrocarbon accumulations (e.g., Manetoe Dolomite). The origin of megacrystalline, white dolomite is the least certain of the three major petrographic types because it has not been observed in recent sediments or in Quaternary rocks. However, the universal association of white dolomite, both as a void-filling and as a replacement mineral, with solution-collapse breccias and solution cavities indicates that it formed after lithification and probably after at least some burial. Several workers have emphasized the common association of lead-zinc mineralization and white dolomite (e.g., Beales and Hardy, 1980; Davis, 1979; Radke and Mathis, 1980) suggesting that both the sulphides and the dolomite precipitated together by means of a single, overall-sulphate reduction reaction at temperatures between 60° and 150°C (Barton, 1967; Radke and Mathis, 1980). But any theory linking the origin of lead-zinc mineralization with the associated white sparry dolomite in spe-
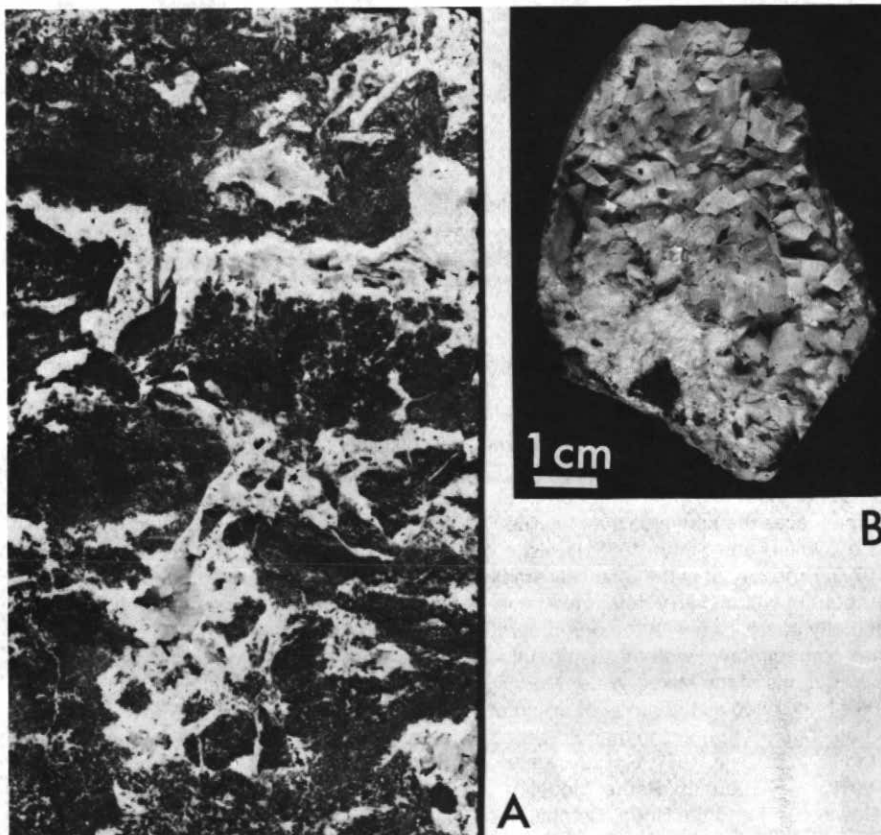

Figure 6 *Megacrystalline white sparry dolomite of the Manetoe Facies (Formation) in the Nahanni Formation of the Yukon Territory. Near centimetre-sized crystals of white sparry dolomite have partly filled spaces created by solution-collapse in the core slab (A). Photograph B, at the same scale, is the exposed inner surface of one of these vugs displaying crystals with markedly curved crystal faces. This type of dolomite is totally fabric destructive. This sample is a core specimen taken 11,707.0 feet below sea level in the gas-producing Columbia et al. Kootaneelee H-38 well.*

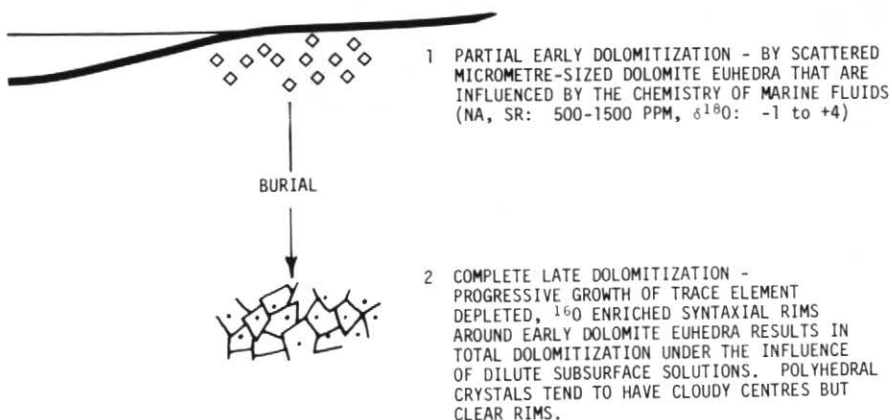THE NATURAL HISTORY OF DOLOMITE CRYSTALS

1   PARTIAL EARLY DOLOMITIZATION - BY SCATTERED MICROMETRE-SIZED DOLOMITE EUHEDRA THAT ARE INFLUENCED BY THE CHEMISTRY OF MARINE FLUIDS (NA, SR: 500-1500 PPM, $\delta^{18}O$: -1 to +4)

BURIAL

2   COMPLETE LATE DOLOMITIZATION - PROGRESSIVE GROWTH OF TRACE ELEMENT DEPLETED, ^{16}O ENRICHED SYNTAXIAL RIMS AROUND EARLY DOLOMITE EUHEDRA RESULTS IN TOTAL DOLOMITIZATION UNDER THE INFLUENCE OF DILUTE SUBSURFACE SOLUTIONS. POLYHEDRAL CRYSTALS TEND TO HAVE CLOUDY CENTRES BUT CLEAR RIMS.

Figure 7 *Many dolomites conform to this type of diagenetic history. The dilute subsurface solutions are probably mixed-water or burial-compaction in origin. It is not known whether trace element depletion in crystal rims could be the result of slow precipitation from more saline solutions of special compositions.*

cific areas must also account for the widespread distribution of dolomite beyond the mineralized area (e.g., Skall, 1975; Gorody, 1980). It must also account for the fact that mineralization in most examples post-dated the emplacement of much of the white dolomite, as, for example in the Pine Point deposit (Skall, 1975) and in the Viburnum Trend of the Mississippi Valley district (Gorody, 1980; Hagni and Trancynger, 1977). The lead-zinc deposits in the St. George Formation in Newfoundland are a notable exception to this generalization (Collins and Smith, 1975). Clearly, the processes responsible for the emplacement of widespread white dolomite masses beyond the mineralized areas are not dependent on the processes of mineralization, although in some places the converse may be true (e.g., Collins and Smith, 1975; Davis, 1977). Probably only the relatively small amount of white sparry dolomite that is actually intergrown with the ore minerals was coprecipitated with these minerals.

Variations of the Mixed-Water Model have been cited as the mode of origin of white sparry dolomites in many recent studies (e.g., Lyle, 1977; Gorody, 1980; Morrow, 1975) but the Reflux Model (Davis, 1977) and the Burial Compaction Model (Griffin, 1965; Jackson and Beales, 1967) have also been cited, in addition to the model based on the late diagenetic sulphate reduction reaction (Radke and Mathis, 1980). Unfortunately, it is evident that little overall agreement exists between workers concerning the origin of megacrystalline white dolomite either in general, or with regard to specific occurrences.

Paragenesis and Texture. Detailed petrographic relationships and textural data in some instances can be used to infer the timing of dolomitization with respect to early events such as the recrystallization of aragonite and high-magnesian calcite (e.g., Schmidt, 1965; Kendall, 1977; Sibley, 1980) or to later events such as the development of stylolites (e.g., Macqueen and Thompson, 1978; Wong and Oldershaw, 1981). Early dolomitization is often controlled by original permeability and the commonly noted preferential dolomitization of finer grained sediments (Murray and Lucia, 1967) is probably due in large part to their great abundance of nucleation sites. Increasingly, multiple episodes of dolomitization are being recognized based mainly on the recognition of a wide variety of petrographic types of dolomite within the threefold petrographic classification outlined above. Many of these varieties have been related to the operation of specific dolomitization models (e.g., Sears and

Lucia, 1980; Mattes and Mountjoy, 1980), so that a given dolostone may have been influenced by the operation of a succession of such models. However, it is not possible at present to relate any of these petrographic types to any particular model in the absence of other data, but the petrographic character of a given dolomite may help to limit the choice of models.

Geochemical Criteria

Geochemical Parameters. Geochemical parameters include compositional variations of the major elements Ca and Mg and the degree of concomitant crystal disorder, variations in the content of minor and trace elements such as Fe, Sr, Na, Mn and other less commonly studied elements (Ba, Si, S, Zn, Pb), and the isotopic composition of carbon and oxygen. A considerable body of literature has arisen in the past two decades dealing with these parameters. The most effective of these studies have combined stratigraphic and petrographic data with geochemical data. Stratigraphic and petrographic data provide a foundation of more readily accessible facts upon which a framework of interpretations involving geochemical data may be built.

Compositional Variations of Major Elements and Crystal Disorder. Compositional variation of major elements in dolomites is commonly expressed as a mole percent of $CaCO_3$ (Fig. 8). Calcium

is almost invariably present in excess of Mg because of the relative ease with which Ca is incorporated into growing dolomite crystals (see Morrow, 1982). There is a growing recognition that the degree of enrichment of Ca in dolostones reflects the Mg/Ca ratio of the precipitational solution (e.g., Füchtbauer and Goldsmith, 1965; Morrow, 1978; Lumsden and Chimahusky, 1980). Typically, dolomicrites or finely crystalline dolostones that are associated with evaporites have Ca contents that are lower than for finely crystalline dolostones not associated with evaporites (i.e., Groups 2 and 3 in Fig. 8). Dolomites in both of these groups are early diagenetic in that dolomitization occurred within the influence of the depositional environment (e.g., Patterson, 1972; Morrow, 1978; and Lumsden and Chimahusky, 1980). The relatively high Mg/Ca ratios of solutions in evaporitic settings (e.g., Reflux and Sabkha Models) induces the formation of nearly stoichiometric dolomites, because the abundance of Mg^{2+} ions compensates for their relative difficulty in precipitating from solution to form magnesium-bearing compounds (see Morrow, 1982). On the other hand, the lower Mg/Ca ratios of solutions in more humid, non-evaporitic settings (e.g., Mixed-Water and Coorong Models) results in the precipitation of more calcium-rich dolomites.

Medium to coarsely crystalline ancient dolomites tend to be more stoichiometric (Group 1 in Fig. 8). One explanation is
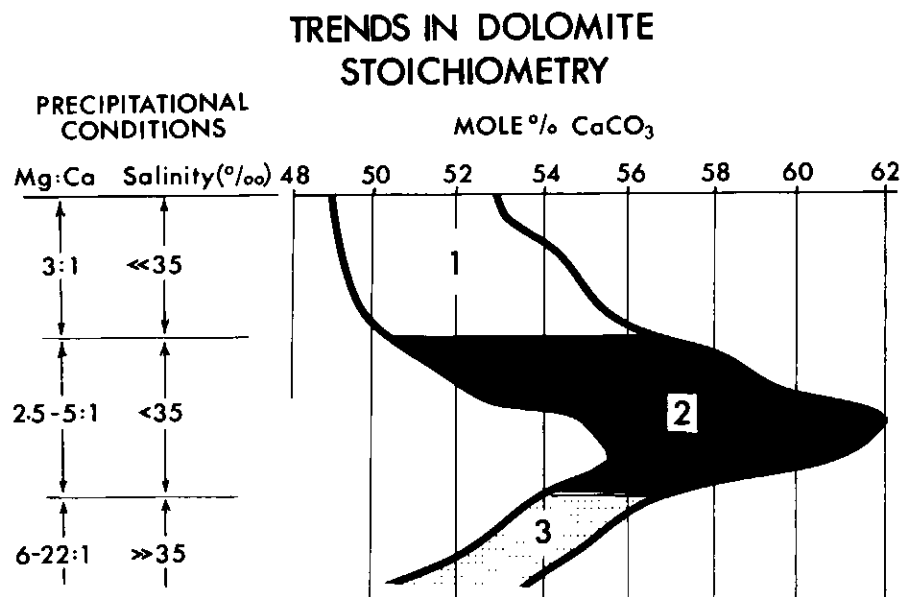

TRENDS IN DOLOMITE STOICHIOMETRY

Figure 8 *A summary diagram incorporating data compiled by Morrow (1978), Mattes and Mountjoy (1980), and Lumsden and Chima-husky (1980) concerning the stoichiometry of dolomite both modern and ancient. Group 1 is composed of ancient sucrosic and sparry dolomites, Group 2 is composed of finely crystalline modern and ancient dolomites not associated with evaporites and Group 3 are finely crystalline modern and ancient dolomites associated with evaporites.*

that scattered, early diagenetic dolomite crystals in incompletely dolomitized limestones continue to grow very slowly in late diagenetic subsurface environments, under the influence of very dilute solutions and possibly elevated temperatures. Slow precipitation may overshadow the influence of low solution Mg/Ca ratios and cause the growth of the trace element depleted, more stoichiometric, coarsely crystalline rims that enclose Ca and trace element-enriched cores of dolomite crystals (e.g., Land et al., 1975; Morrow, 1978). The net effect of continued crystal growth during later diagenesis is the formation of dolostones whose bulk composition approaches that of stoichiometric dolomite (e.g., Fritz and Jackson, 1971 and Fig. 7). Certainly this interpretation explains the depletion in most trace elements that invariably occurs towards the periphery of dolomite crystals and implies that they are the products of variants of the Mixed-Water or the Burial-Compaction Models.

Some data concerning the departure from ideal cation ordering within dolomite crystals has been gathered, particularly for dolomites forming in modern environments (e.g., Patterson, 1972). Unfortunately few ancient dolostones have been analyzed for this parameter. Comparisons of the degree of cation ordering as determined by the analysis of superstructure reflections on x-ray patterns with the degree of Ca enrichment might impart some insight into how closely these parametres are correlated. For example, crystal disorder not associated with Ca enrichment would reinforce the conclusion that finely crystalline, stoichiometric dolomite was rapidly precipitated from saline solutions with a high Mg/Ca ratio.

Trace Element Variations. The concentration of trace elements in dolomite ideally provides a means for estimating the composition of the precipitational medium because the fractionation of an element, such as Sr, from a liquid phase into a solid phase is proportional to its concentration in the liquid phase (amongst a variety of other factors). This implies that dolostones with greater concentrations of trace elements, such as strontium or sodium; precipitated from more saline solutions; but quantitative interpretation of solution compositions based on dolomite trace element data are not feasible at present. Estimates of low temperature partition coefficients based on high temperature experiments (Katz and Matthews, 1977; Jacobson and Usdowski, 1976) or derived from element concentrations in Holocene dolomites

(Patterson, 1972; Land and Hoops, 1973; Behrens and Land, 1972) have not been satisfactory (Land, 1980; Veizer et al., 1978) and direct determinations from precipitation experiments are of course not possible at reasonable temperatures.

The extreme difference in the contents of trace elements in ancient versus modern dolomites is the salient fact to emerge from trace element studies. The two most commonly studied elements, Sr and Na, are present in concentrations approaching, or greater than, 1000 ppm in modern dolomite but in ancient dolomites, they commonly are less than a few hundred ppm (Land et al., 1975; Mattes and Mountjoy, 1980). Other elements such as S, K, Al, Ba, Zn, Mn and Fe have been analyzed only sporadically in ancient dolostones and, little, if any data exists for modern dolomites.

The phenomenon of intracrystal variations or zonations with respect to trace elements within ancient dolomite crystals is commonly observed, even in crystals smaller than 10 μm (e.g., Choquette and Steinen, 1980). Studies of element zonations in dolomite crystals have also concentrated on the elements Na and Sr, and in addition, there is some data for Fe and Mn. A general pattern exists of high Na and Sr contents in the cores surrounded by rims with much lower contents. (Fritz and Katz, 1972; and Fig. 7). On the other hand, Fe, and possibly also Mn, tend to exhibit an opposite zonation with higher concentrations toward the periphery of crystals. Bulk sample analyses, of course, are weighted averages of these zoned crystals. Some studies have shown slight statistical differences in the bulk trace element contents of a variety of dolostone types (Veizer et al., 1978).

Any explanation of dolomitization must account for both the element zonation of ancient dolomite crystals, and their reduced trace element contents, particularly for the aphanocrystalline and fine to medium crystalline, sucrosic replacement dolomites that commonly have cloudy centres and clear rims. It is difficult to avoid the conclusion that as dolomite crystal growth continues, it does so under the influence of progressively more dilute solutions that dissolve pre-existing calcite and precipitate dolomite. It seems likely that the cores of ancient dolomite crystals, in largely or totally dolomitized rocks, represent individual crystals that formed in shallow early diagenetic environments under the influence of seawater or modified seawater solutions. Subsequent burial of these rocks into deeper subsurface environments may have been accompanied by the continued precipitation from more dilute solutions of dolo-

mite overgrowths that are relatively inclusion-free, more stoichiometric and lower in Sr and Na (Fig. 7). Such dilute and subsurface solutions are consistent with the operation of either the Mixed-Water Model or the Burial-Compaction Model. Ion filtration during burial compaction of shales can yield dilute solutions that may participate in the dolomitization of nearby carbonates, although there are limitations on the potential amount of dolomite that can be formed.

The commonly observed high Fe concentration of up to several mole per cent in the rims of larger crystals, particularly in those that have precipitated as open space fillings with complex zonation (as revealed by staining and cathodoluminescence) have been related qualitatively to diagenesis in the reducing subsurface phreatic zone, with fluctuations in the supply of Fe^{2+} (Wong and Oldershaw, 1981; Gorody, 1980). These observations are also consistent with either a Mixed-Water or Burial-Compaction Model.

Coarsely crystalline, ferroan and nonferroan, white, sparry dolomites directly associated with Pb-Zn mineralization or with hydrocarbons have also been interpreted to have been precipitated from saline host fluids, based primarily on the occurrence of very saline fluid inclusions in associated minerals (Radke and Mathis, 1980). Certainly subsurface precipitation of dolomite at the inferred temperature of greater 100° C is less subject to kinetic inhibitions. However, it is puzzling that the few occurrences of white, sparry dolomite associated with Pb-Zn mineralization which have been analyzed systematically for their trace element contents (e.g., Fritz and Jackson, 1971) are characterized by low trace element contents in conformance with the general rule regarding trace elements in dolostones. Saline solutions should precipitate dolomites markedly enriched in trace elements. This major contradiction concerning the origin of white megacrystalline, sparry dolomite cannot fail to stimulate further research, and trace element studies will undoubtedly play a key role in resolving this paradox. Distribution coefficients for some elements, such as strontium (Jacobson and Usdowski, 1976) have been determined for the high temperature (>100° C) precipitation of dolomite which might enable quantitative modelling of trace element distributions.

Trends of trace element concentrations within dolostone bodies have also been used as indicators of the direction of fluid migration during diagenesis (Land, 1980). This type of data may allow discrimination between models for dolomitization

that require different directions of fluid migration during dolomitization, such as the Burial-Compaction Model versus the Mixed-Water Model.

Oxygen and Carbon Isotopes. Land (1980) has recently summarized the status of stable isotope studies (δ^{18}O, δ^{13}C) of dolomite and has pointed out the considerable difficulties in their interpretation. Qualitative interpretation of oxygen and carbon isotope data involves an appraisal of the influence of the variables of temperature and solution compositions. Higher temperatures and dilution by meteoric water favour the incorporation of the light ^{16}O isotope into the solid phase so that dolomites that are greatly enriched in ^{16}O may have been influenced by high subsurface temperatures (Mattes and Mountjoy, 1980) or by dilute ^{16}O rich groundwater (Land *et al.*, 1975). The interplay of these factors is complicated by the probable existence of secular changes in the isotopic composition of ancient seawater (Veizer and Hoefs, 1976).

One certainty with regard to oxygen isotopes in ancient dolostones is that their bulk δ^{18}O is much lower than that of modern dolomites (Land, 1980). Probably dolomite crystals are zoned with respect to their ^{16}O content in a manner analogous to that described for trace elements. Although intracrystal variations in ^{16}O have not been described, there does appear to be a correlation between increasing crystal size and decreasing δ^{18}O values for dolomite crystals (Fritz and Jackson, 1971; Land *et al.*, 1975; Mattes and Mountjoy, 1980) which is consistent with the peripheral enrichment of ^{16}O. This may corroborate the interpretation that the growth of dolomite crystals continues under the influence of progressively more dilute solutions. Regional trends of δ^{18}O within dolostones may also corroborate trends in their trace element distributions (Land *et al.*, 1975).

In contrast to oxygen isotopes, the ratio of stable carbon isotopes (δ^{13}C) in most ancient dolostones is virtually the same as in Holocene dolomites with perhaps only a slight tendency for ancient dolostones to be enriched in ^{12}C (Land, 1980). This may simply reflect the fact that the amount of carbon in dissolved species in natural solutions is very small compared to the amount of carbon in the carbonate rocks that are being replaced. There are a few dolostones enriched in ^{12}C that may have been involved in bacterial reduction reactions.

Summary and Conclusions

The description and classification of dolostones previous to about 1960 was a comparatively simple matter as dolostones were commonly classified as 'primary' or 'secondary' depending primarily on whether the sedimentary fabric of the rock was preserved or destroyed by dolomitization. The additional category of 'hydrothermal' dolomite was used and is still used (Jones, 1980) occasionally to characterize masses of coarsely crystalline, space-filling white dolomite. Little was understood concerning processes of dolomitization and consequently most dolostones were subjected to comparatively casual investigations in spite of their economic importance.

The decades following 1960 were marked by the discovery of many examples of modern dolomitization in a variety of environmental settings (e.g., sabkha, ephemeral lake, deep carbonate aquifer) and the characterization of the chemistry of these environments has generated a greatly increased understanding of the chemistry of dolomitization. At the same time, new analytical techniques (e.g., x-ray fluorescence, electron microprobe, etc.) have permitted a better overall characterization of the mineral dolomite itself in both ancient rocks and modern occurrences. The rapid increase in knowledge of other aspects of geoscience such as fluid dynamics and chemical changes during the burial compaction of sediments and the hydrology and chemistry of coastal aquifers have had a profound influence on the development of theories concerning dolomitization. This multidirectional increase in knowledge has resulted in the development of a variety of dolomitization models and an increase in the number of criteria available to discriminate between models.

The objectives of this review have been twofold. First, to outline clearly the models for dolomitization that have arisen and to review the criteria commonly employed in choosing between these models. The multitude of models for dolomitization that are now recognized are moderately to well defined. It seems evident to this reviewer that variants of the Mixed-Water Model are the most widely applicable — the first among equals. This is not to say that the Mixed-Water Model in itself constitutes the answer to the 'dolomite problem' only that many ancient dolostones may have formed by the operation of this model. The other models are certainly viable but perhaps not to the same degree and in less regional settings. The importance of a model such as the Burial-Compaction Model lies not in its potential for forming

vast amounts of dolomite but rather in its capacity for modifying pre-existing porosity.

Unfortunately the criteria for choosing between models are not as definitive as could be desired and many of these criteria have been interpreted in more than one way. Indeed it could be argued that the same criteria in some dolostones such as the Presqu'ile Formation (or facies) have been used to support a succession of different models as these models successively rose in popular esteem. Hopefully, the time of fads is past and the choice of a model for dolomitization by present day workers is based on an equitable consideration of all available criteria and by practicing the spirit embodied in the phase "multiple working hypothesis." Obviously much work remains to be done.

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