
CONTROLS OF KARST DEVELOPMENT

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ABSTRACT

Many variables other than the carbonate and hydro-carbonate dissolution of limestone contribute to the development of karst landscapes. Atmospheric CO₂, previously thought to be the most important factor in karst evolution, has recently been relegated to a much less significant role. Biologic processes (bacterial decay...) and organic acids (humic...) have been shown to be as effective as any agents in promoting karstification. Small amounts of MgCO₃ in a limestone greatly inhibit dissolution, yet other contaminants such as iron oxide, clay minerals, and silica seem to have little effect. The disproportionately high occurrence of karst in Mesozoic rocks may be due to a combination of their high strength from compaction with time and high degree of "secondary permeability" caused by tectonic stresses. In general, a pure, massive, holocrystalline limestone with a high degree of jointing is considered favorable for the development of karst.

INTRODUCTION

In the interest of simplicity, lithologic variation, other than that within the common term "limestone", has been omitted. This is not to suggest that karstification doesn't proceed in areas composed predominantly of dolomite, granite, andesite, and other rocks. By concentrating on the major processes which control karst evolution, it has not been feasible to include a description of the resulting landforms or of any hydrologic processes. Cave formation, considered by some to be a karstic phenomenon has, do to the conflicting nature of its mechanisms of development, been left untreated. In general only those processes considered important for the development of karst in limestone (dissolution, the origin of CO₂ and its absorption in water, biogenic agents, organic and inorganic acids) along with lithostructural properties of limestone and their impact on karst evolution are discussed.

DISSOLUTION OF LIMESTONE

(Bögli 1960) describes 4 chemical phases responsible for limestone dissolution. In the first phase, the limestone dissolves directly in water without influence of any other agents:



Table 1. gives values of calcite content (in mg/l) for water in equilibrium, at various temperatures. Bögli describes this ionic reaction as being very fast, estimating about one second as the time required. (Gerstenhauer and Pfeffer 1966), however, through laboratory experiments, found this process to take nearly a full day, even when using water which contained some CO₂ (which accelerates dissolution).

In the second and third phases, the dissolved CO₂ reacts; only a small percentage (.7%) being in the form of carbonic acid. The remaining 99.3% is physically dissolved (Pia 1953). In the second phase, the H[±] ion from the carbonic acid combines with the CO₂ ion of the first phase:



Because of the association of these two ions, CO₃ is thrown out of

Table 1.

Temperature °C	Dissolved CaCO ₃ (mg/l)	Author (year)
8.7	10.0	Schloesing (1872)
16.0	13.1	Schloesing (1872)
16.0	16.0	Trombe (1952)
25.0	14.3	Schloesing (1872)
25.0	14.33	Schoeller (1956)
50.0	15.04	Schoeller (1956)

solution equilibrium and renewed solution of limestone results. The balance between the chemically and physically dissolved CO₂ is disturbed as well. In phase three, the physically dissolved CO₂ forms carbonic acid and its ions:



This requires about one minute. It is the beginning of the chain of reactions, at the end of which, further solution will occur. The total amount of limestone dissolved in stages two and three depends upon the original CO₂ content of the water. Assuming a constant temperature, the CO₂ content of the water is a linear function of the PCO₂ (partial pressure) of the air. Table 2. shows the amount of CO₂ in water given the PCO₂ in the air and its temperature. Along with equation 1, the amount of lime (in mg/l) soluble in the original precipitation can be calculated.

$$\text{Eq. 1.) lime in mg/l} = \frac{100}{44} \cdot \text{CO}_2 \text{ (mg/l)}$$

Table 2.

p in the air	0°C	10°C	17°C	25°C	30°C
0.0001	0.34	0.28	0.19	0.15	0.13
0.0003	1.01	0.70	0.56	0.45	0.39
0.001	3.36	2.34	1.88	1.49	1.31
0.0025	8.40	5.85	4.70	3.73	3.28
0.005	16.8	11.7	9.4	7.46	6.56
0.0075	25.2	17.6	14.1	11.2	9.8
0.01	33.6	23.5	18.8	14.9	13.1
0.05	168	117	94	74.6	65.3
0.1	336	235	188	149	131

(after Bögli 1960)

The fourth stage involves the absorption of more CO_2 from the atmosphere and doesn't begin until the first three have finished. Bogli estimates between one day and 60 hours as the time required for this final equilibrium to be reached.

Mixing Corrosion

When two masses of water, each with different PCO_2 's and both saturated with respect to CaCO_3 mix, renewed aggressiveness may result due to the mixing effect. It can be seen from Fig. 1. that this is due to the curved nature of the solubility graph, in comparison to a linear increase (or decrease) in both calcite concentration and PCO_2 upon mixing. Although maximum undersaturation per unit water is greatest for equal amounts of low and high PCO_2 waters (Bogli 1964); total undersaturation (undersaturation per unit volume x water) is greatest when largest amounts of low PCO_2 waters are in mixture (Thraikill 1968).

Thraikill points out four common circumstances which may defeat the mixing effect where it would normally occur. (For purposes of these circumstances assume that vadose water (that above the water table) is the mass high in PCO_2 relative to phreatic water (that water in the zone of saturation)).

1. Vadose waters may be in equilibrium with a low PCO_2 . Although PCO_2 's of vadose waters are usually considerably higher than those of the atmosphere, it seems likely that flows representing captured surface streams, which descend to the water table quickly, would have PCO_2 's not much higher than the atmosphere, if at all.

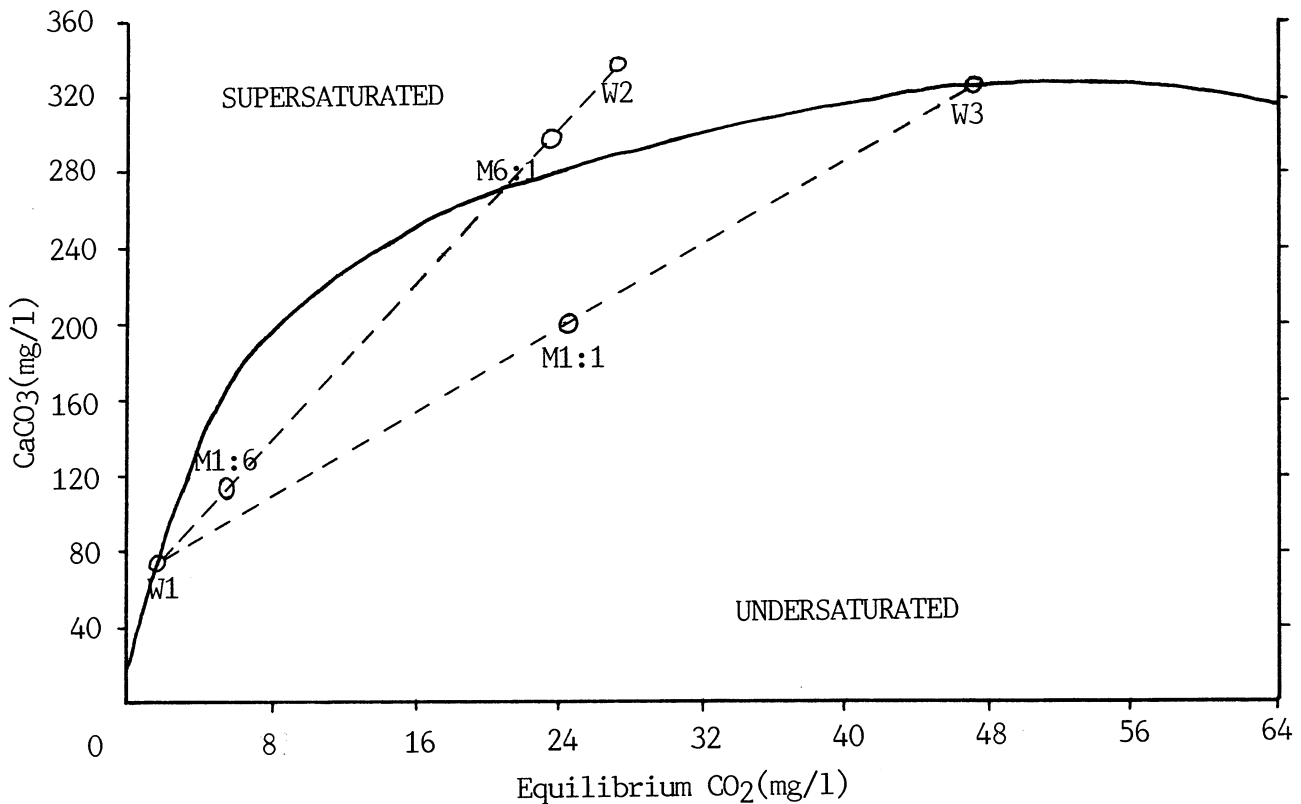


Fig.1. The mixing effect. Two waters, saturated with respect to CaCO_3 may, upon mixing, result in an undersaturated mixture, as is the case with W1 and W3. If, as in the case of W1 and W3, one of the waters is supersaturated, the resultant mixture may be either undersaturated or supersaturated, depending on the ratio of quantities mixed. (after Jennings, from Bögli 1964 and Thrailkill 1968).

2. Phreatic waters may be in equilibrium with a high PCO_2 . It is difficult to obtain an accurate measurement of the pH of phreatic waters (necessary for a PCO_2 measurement) because laboratory measurements of pH are made some time after collection and are not representative of in situ pH conditions (Back 1963). However, data from G.W. Moore (in Hostetler 1964) suggests that an equilibrium PCO_2 higher than that of the atmosphere may exist in some ground-water bodies.
3. Vadose water may be supersaturated. The deposition of calcite is often not as rapid as the outgassing of CO_2 , thus supersaturation results.

4. Ground-water derived from vadose seepage could be supersaturated in the same manner as above.

Bögli indicates that corrosion by mixed waters occurs only in underground water channels, but (Gams 1965) points out that this process could just as easily work in surface streams and rivers.

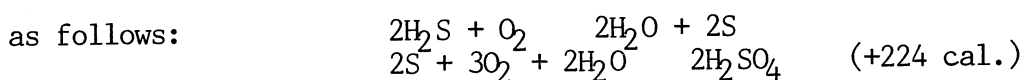
Other Agents of Limestone Corrosion

Biogenic agents, both for their contribution to CO₂ in karst waters and their production of organic acids through decay, have a tremendous impact upon karst development. The partial pressure of CO₂ in the normal atmosphere is 3×10^{-4} atm. It is believed that the amount of CO₂ in the ground air is 10 to 100 times as great (Bogli 1960); depending upon the degree of humus decay and the amount of carbonic acid given off by roots. These being dependent on climate, it is easy to see why karstification in tropical zones proceeds much quicker than in polar zones despite the capacity of cold water to absorb much more CO₂ from the atmosphere than warm water.

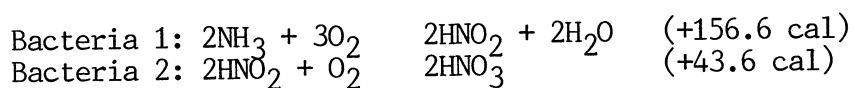
The decay of organic matter and the metabolic processes of microorganisms produce the following chemical agencies important in limestone corrosion: formic acid, oxalic acid, acetic acid, propionic acid, lactic acid, secretions of various roots, and carbonic acid (Jakucs 1977). All of these agents liberate Ca salts upon interaction with limestone. Jakucs lists fulvic and crenic acids as the most important corrosive organic acids produced by the decay of plants and animals. Humic and huminic acids are also important in limestone corrosion; although their reactant products tend to preci-

pitate in acidic mediums.

Inorganic acids and salts resulting from the biochemical or inorganic processes of weathering can be important in the corrosion of limestone. Two of the most common, sulphuric and nitric, may exist in the soil as a result of several processes. Sulphuric acid is usually a product of the oxidation of sulphides (e.g. pyrite) and hydrogen sulfide (H₂S). The H₂S compounds may result from the decay of living organisms and/or the chemical decomposition of certain inorganic soil constituents. The formation of sulphuric acid can be accomplished, according to (Feher 1954), by the genera of sulphur bacteria *Achromatium*, *Beggiatoa*, *Thiotrix*, and *Thioploca*. The bacteria split off sulphur from the H₂S molecule, store it in the form of droplets in their plasma, and then burn it to derive energy for the assimilation of carbon. In simplified form, the process operates



Nitric acid in the soil originates most often from ammonia, usually present as a result of organic decay. Ammonia is oxidized into corrosive acids by two groups of collaborating bacteria. (Vinogradsky 1892) showed the nitrification of the soil due to these bacteria to proceed in the following way:



Nitric acid may also occur in the soil from the absorption of nitric acid from the atmosphere produced by electrical discharges during thunderstorms (Reiter 1960).

(Bloom 1978) mentions that phosphatic mixtures, such as bat guano, serve as strong limestone corrosives.

CLIMATIC CONTROL OF KARST DEVELOPMENT

Classical ideas about karst came primarily from the Dinaric karst of Yugoslavia and the karst of Central Europe. Differences between these regions came to be explained by the theory of a geographical cycle; assigning terms such as young, mature, and old to various karst regions based on the landforms present in each. Ideas about a karst cycle closely parallel those of landform evolution with respect to river valleys brought forth by Davis in 1899. (Grund 1914) defines a young karst as one in which dolines have developed, but some of the original surface is preserved between the dolines. He defines a mature karst as beginning when the karst of the original surface is destroyed due to doline enlargement, and suggests the "cockpit" karst of Java and Jamaica to be representative of this stage. Further maturing, lowering the surface to the corrosion base level (karst water table), suggests Grund, is the beginning of old age. Depending on the homogeneity of the rock, a new landscape is formed. Given our present state of knowledge, it is no longer acceptable to suggest that karsts as different as those in Java and in Central Europe have evolved under the same mechanisms, differing only in their stages of development.

It has been known for some time that the absorptive capacity of water with respect to CO₂, and thus the corrosive ability of that water, varies inversely with temperature. Knowing this, Corbel (1954, 1955, 1959) a famous French karst scientist, attempted to prove that karstification in polar regions proceeded at a greater rate than that of tropical regions. He showed that the water of rivers draining polar

karst regions contained about 10 times as much dissolved CaCO_3 as the water from rivers draining tropical karst regions. This seemed to support his theory. Corbel based his conclusions on only one thing: the amount of dissolved CaCO_3 in the drainage water.

It is now known that many factors govern the dissolution of limestone.

1) (Jakucs 1977) states that the soil, rich in decaying organic matter, may contain 100 times more CO_2 than the atmosphere. Infiltrating water is in contact with the soil over a large area and for an extended period of time, allowing saturation equilibrium to be attained.

2) Even the same soil varies in its CO_2 content. (Trombe 1951, 1952) has shown the rendzina soils of the French karsts to contain up to 10% CO_2 in the summer, but practically none in the winter; just as the meagre soils, without vegetation, and on high mountains contain none. The atmosphere and dry soil both contain .03% CO_2 , whereas wet soils, like those found in the tropics contain as much as 20% CO_2 (Monroe 1966, Harrison and Subramania Aiyer 1913, 1916).

3) (Jakucs 1977) states that other organic and inorganic acids are just as effective as carbonic acid in corroding limestone: ((Monroe 1966) believes that while HNO_3 and humic acid may cause corrosion; most acids are transformed to CO_2 by bacteria almost as soon as they form).

Even if Corbel had included these other agents, basing his conclusion on the amount of dissolved CaCO_3 in the drainage water would have led to substantial errors due to the lack of consideration of that portion of the water eventually absorbed by vegetation from the soil and being recycled by evapotranspiration; as well as that fraction of the water lost to direct evaporation. These fractions are greater the

hotter and wetter the climate (Jakucs 1977). The dissolution potential of rain water is not decided, of course, only by the percentage drained by the rivers. The reversibility of hydrocarbonate dissolution is another neglected factor. The water draining a karst region could, upon warming, or lowering of PCO_2 , allow escape of CO_2 , thus causing the precipitation of $CaCO_3$. If this had occurred, an analysis of drainage waters wouldn't give any clues as to the amount of $CaCO_3$ in solution at one time.

Taking these factors into account, it is easy to see why karstification in humid, tropical regions proceeds at a rate 10 times that of polar regions; not less intense by a factor of 10, as Corbel suggested considering the $CaCO_3$ amount of the drainage rivers exclusively. Jakucs has formulated an important thesis of karst morphology: "cold karsts evolve towards leached skeletal karsts, whereas, owing to the precipitation (either at deeper levels or almost in situ) of the calcium carbonate dissolved at higher levels, the karsts in hot climates develop into massive karsts, with calcium carbonate transport largely vertical, and the horizontal component severely limited". This is why polar karsts exhibit no deposits of calcareous tufa and their caverns are so poor in dripstones (Tell 1962, Rohdenberg and Mayer 1963).

Figure 2. shows the relative intensities of karst corrosion (double line) between various climatic zones, and the percentage contribution of individual agents within that zone. In this figure, as in Table 3. and Figure 3. the letter designations are as follows:
A - High mountain and periglacial

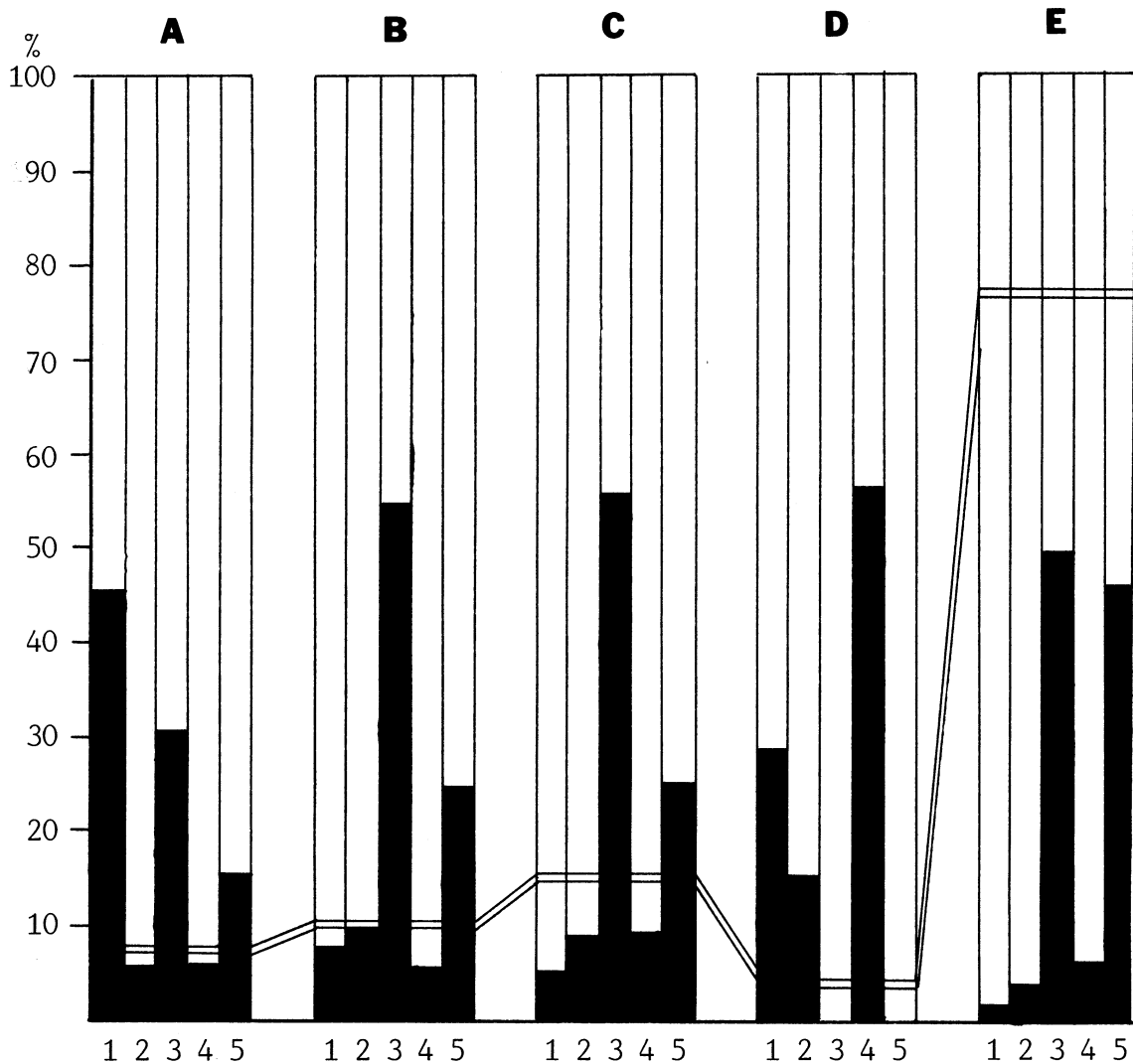


Fig.3. Relative intensities of karst corrosion and percentage contributions by various agencies in the fundamental climatic zones of karst morphology (after Jakucs 1977).

Agencies of karst corrosion:

- 1 - CO₂ fraction of atmospheric origin; 2 - idem, due to inorganic pedogenic processes (e.g. weathering); 3 - idem, due to biogenic processes of pedogenesis; 4 - contribution of other inorganic acids; 5 - contribution of organic acids (humic, huminic, and root acids)

B - Temperate

C - Mediterranean

D - Desert

E - Tropical

Only the percentage contribution of the individual agencies within each climatic zone is indicated, thus all comparisons must be made between the agencies within one particular zone, indicating nothing about its difference in magnitude from one zone to another. For example, while the contribution of inorganic acids for both temperate and tropical zones is about 5%, the magnitude of corrosion by these acids is much higher in the tropics because the total corrosion is much higher (by a ratio of 72:9).

Table 3. shows the absolute intensities of the various agents responsible for karst corrosion. These figures were obtained by considering the relative percentage contribution of an agent within its climatic zone, and the percentage believed to be contributed by that zone to total world-wide karstification. Figure 4. is the graphical representation of Table 3. Curve 1 (atmospheric CO₂) is opposed to the behavior of the other curves. This can be explained by the Henry-Dalton law of gas absorption, i.e. that cold water will absorb more gas than warm water, other conditions being equal. Also, a disproportionate decrease in intensity can be observed between zone A (mountain & periglacial) and zone B (Mediterranean) considering the mild increase in temperature. A greater increase in temperature is found between zone B and zone E (tropics), yet the decrease in

Table 3.

	A	B	C	D	E	W
Atmospheric CO ₂	2.70	0.63	0.48	0.30	0.36	4.47
CO ₂ of inorganic origin	0.30	0.81	0.96	0.15	1.80	4.02
Biogenic CO ₂	1.80	4.86	6.60	0.00	36.00	49.26
Inorganic acids	0.30	0.45	0.96	0.55	2.88	5.14
Organic acids	0.90	2.25	3.00	0.00	30.96	37.11
Total	6.00	9.00	12.00	1.00	72.00	100.00

(after Jakucs 1977)

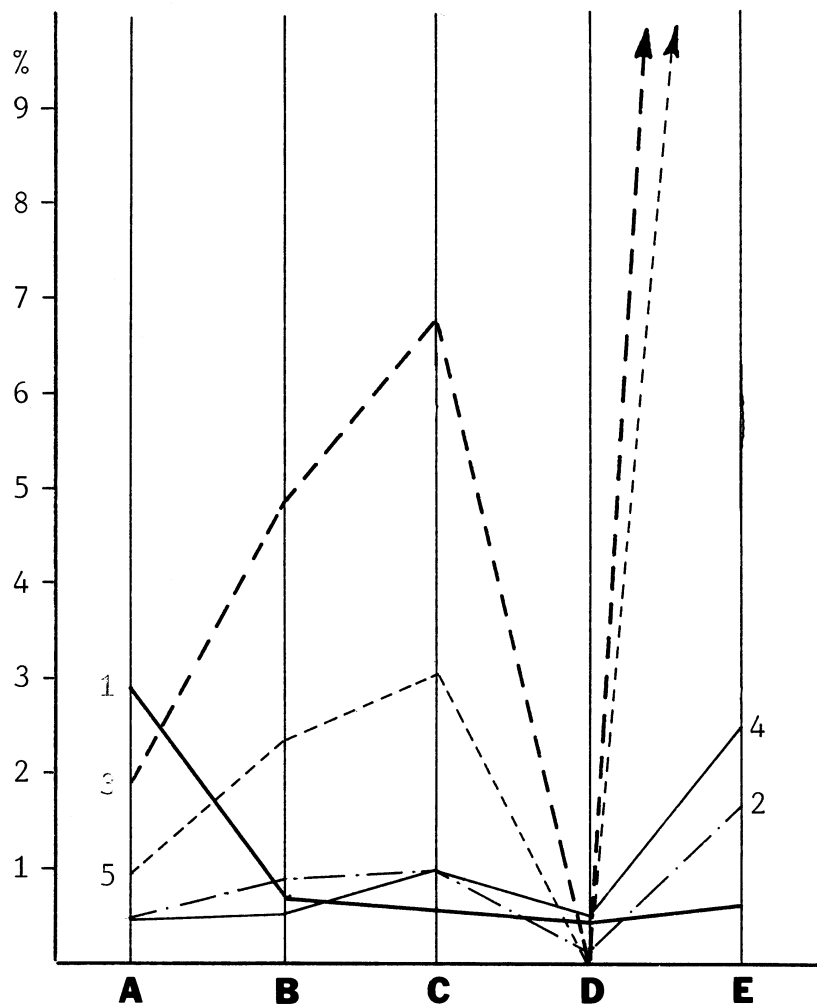


Fig. 4. Absolute intensities of karst agents. (after Jakucs 1977)

intensity of the atmospheric contribution to CO₂ between the two zones is much less. This apparent discrepancy is attributed, by Jakucs, as due to the fact that in zone A most of the precipitation is in the form of snow and drizzle which are both favorable for gas absorption. This is particularly true for snow, which is in contact with the atmosphere for an extended time. Since the slight increase in intensity between D (desert) and E cannot be attributed to a decrease in temperature, it is probably due to the higher overall CO₂ level of

the tropical atmosphere. It is obvious that atmospheric CO₂, to which much importance has been ascribed in the past, is important only in periglacial and mountain climates, and is negligible in others (the biogenic factor is 100 times as important in the tropics). On the other hand, it is obvious that curve 3 (biogenic CO₂) and curve 5 (organic acids) are very sensitive to climate, are the most important factors in karst corrosion for the earth's surface, and are more important the greater the biomass on the ground surface.

INFLUENCE OF LITHOLOGY AND STRUCTURE

In a previous section, hydrocarbonate dissolution of limestone was discussed. While it is important to understand the mechanisms involved in limestone dissolution, it should be realized that rocks falling under the common name "limestone" rarely approximate the 100% calcite composition assumed in laboratory experiments, thus considerable variations arise in karst development, both quantitatively and qualitatively because of these variations.

The most common accessory constituent of limestone is magnesium carbonate. Iron oxide, clay minerals, sand grains, and inclusions of silica gel are other common constituents. These contaminations are, as a rule, insoluble in ground water and karst water, leaving sizeable accumulations of residue during karst genesis, thereby playing a controlling role in karstification (Jakucs 1977). Table 4. lists various types of limestone and their chemical compositions.

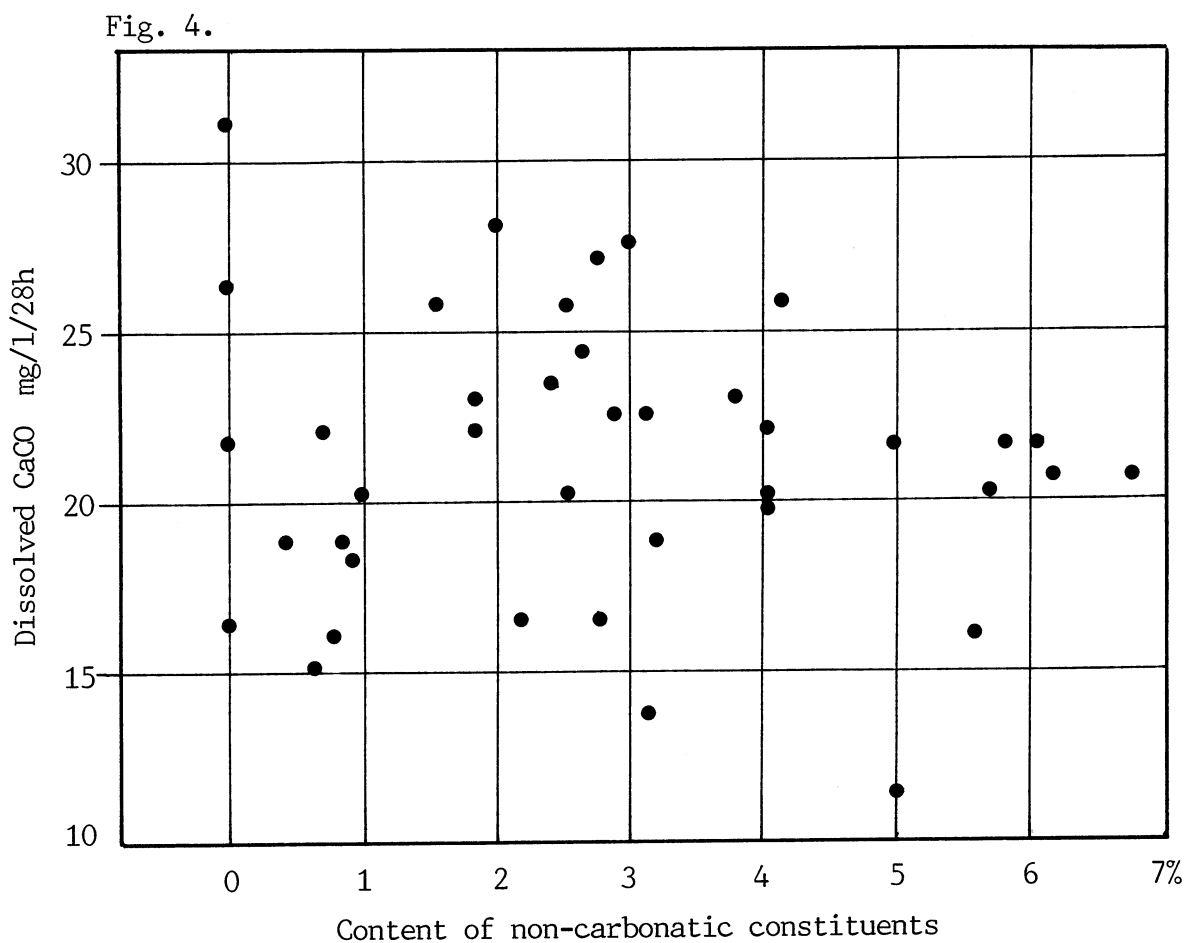
(Mandy 1954) investigated solubilities of different limestones and concluded what had been supposed for many years; dolomite (and high Mg limestone) is much less soluble than any pure limestone. Furthermore, experiments by Gerstenhauer and Pfeffer show that solubilities of limestone depend more upon the $MgCO_3$ content of that rock than on its $CaCO_3$ content. As the $MgCO_3$ content increases, the decrease in solubility is disproportionately small; concentrations of $MgCO_3$ higher than 5% do not represent any further solubility decrease over concentrations around 5%. Gerstenhauer and Pfeffer also carried out these experiments using calcium and magnesium carbonate

Table 4.

Sample designation	% CaO	CO ₂	MgO	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	TiO ₂	MnO	K ₂ O	Na ₂ O	P ₂ O ₅	SO ₃	Cl
Recent Corallian limestone (after Clarke)	55.2	43.7	0.2	0.2	*	*	-	-	-	-	-	-	-
Calc. tufa from Ivanovo- Voznesensk (after Shvetsov)	54.3	44.5	0.2	0.4	0.7	*	-	-	-	-	-	-	-
Aver. comp. of 345 lime- stone samples (aft. Clarke)	42.6	41.6	7.9	5.2	0.8	0.5	0.1	0.1	0.3	0.1	0.1	0.1	*
Aver. comp. of 498 lime- stone samples (aft. Clarke)	40.6	35.6	4.5	14.1	1.8	0.8	0.1	*	0.6	0.6	0.4	0.1	*
Cretaceous limestone (after Rengarten)	50.1	39.8	0.4	5.7	2.4	0.2	-	-	-	-	-	-	-
Cretaceous limestone (after Dobrov)	52.4	43.6	0.5	1.0	0.9	-	-	-	-	-	-	*	-
Liassic limestone (after Gedeon)	53.1	42.8	*	0.3	3.6	*	-	-	-	-	-	-	-
Russet calcaire ammonitico (after Blanck and Weisse)	52.8	41.8	0.3	0.4	0.8	0.1	-	-	-	-	-	-	-
Triassic limestone (after Jakucs)	53.2	44.0	2.0	0.6	0.1	0.1	-	*	-	-	-	-	-
Lower Carboniferous lime- stone (after Shvetsov)	55.4	44.1	*	0.1	0.1	0.1	-	-	-	-	*	-	-
Carboniferous cherty lime- stone (after Shvetsov)	41.2	32.3	*	26.4	0.2	*	-	-	-	-	-	-	-
Pink calcaire ammonitico (after Blanck and Weisse)	55.1	43.0	*	0.2	0.4	0.1	-	-	-	-	-	-	-

* = in traces only

powders of analytical purity. Again, it was shown that small amounts of $MgCO_3$ significantly decrease the solubility of the mixture, and that further increases in $MgCO_3$ entail much less than the proportional reduction in solubility. However, it was also noted that the solubilities of natural limestones are much higher than those of the powder with a corresponding Ca:Mg ratio. Presumably, this difference is due either to the non-carbonate contaminants promoting solubility in natural limestone, or to the influence of crystallinity and texture. Jakucs, in order to determine the cause of these solubility differences has, using the data of Gerstenhauer and Pfeffer, plotted the percentage of non-carbonatic constituents against $CaCO_3$ dissolved over a



28 hour period (solubility) for the 46 samples used. Figure 4 indicates no particular relationship between these two variables. This seems to indicate lithostructural differences as the reason for solubility differences.

Four of the samples used by Gerstenhauer and Pfeffer were composed entirely of Ca and Mg carbonate, containing no other contaminants. Discounting lithostructural characteristics of these samples, their solubilities should reflect the Ca:Mg content exclusively. Plotting these points on a graph of solubility vs. Ca:Mg ratio (from Gerstenhauer and Pfeffer using Ca/Mg powder) should reveal a location coincident with the graph obtained from the limestone samples. Fig. 5 shows the actual case to be quite different, again indicating lithostructural differences as the cause of solubility contrasts.

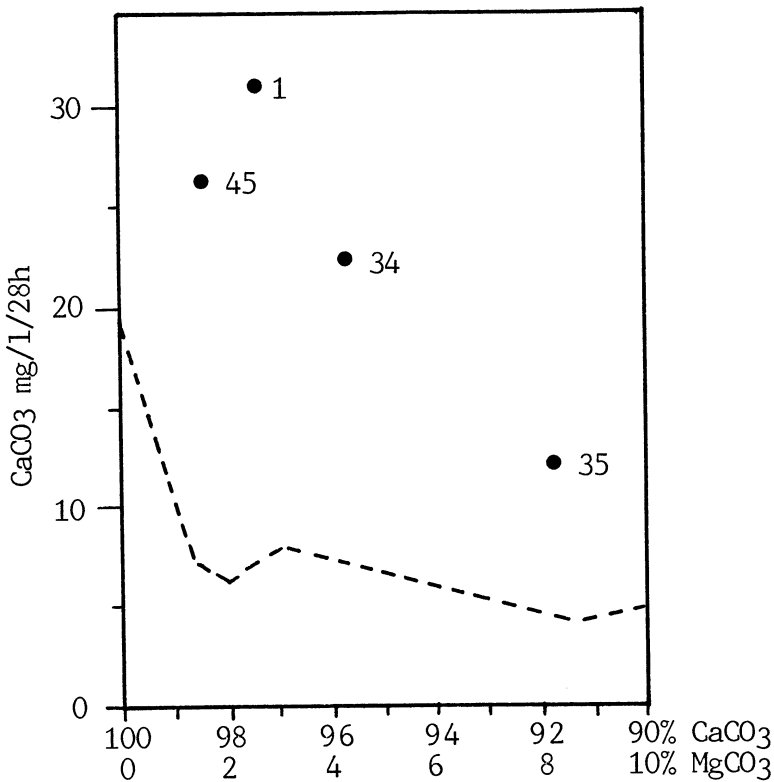


Fig. 5. (after Jakucs)

Lithostructure and Crystallinity

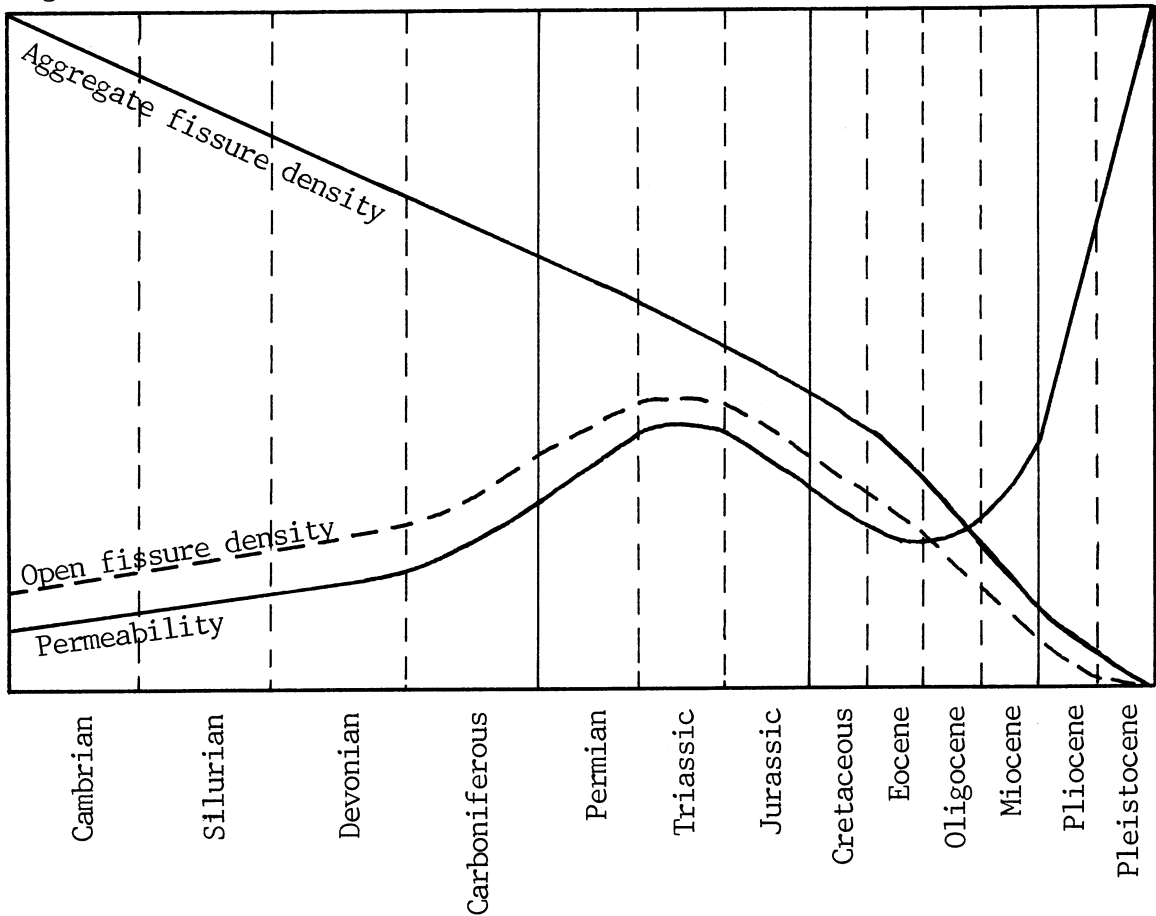
The texture and lithostructure of a limestone can be described by many different parameters: crystallinity, porosity, degree of fissuration, and type and extent of bedding, among others. All these interact to influence the solubility of the rock.

The mechanical strength of limestone greatly affects the development of karst in that region, and is closely related to its porosity and consolidation. Weak limestones may collapse upon solution of underlying rock, affecting surface slope, altering the hydrology and inhibiting cave development. It is in relatively strong limestone, therefore, that typical karst features are observed most often.

Permeability is one of the most important prerequisites for karst evolution. Due to compaction and cementation, however, the stronger limestones tend to have less primary permeability than weaker limestones. Infiltration occurs in strong massive limestones by means of secondary permeability. Secondary permeability develops along planes of weakness, through solutional enlargement (Jennings 1971). The relationship between permeability, fissure density (open and aggregate), and geologic time is plotted in Fig.6. Aggregate fissure density (open fissures plus those closed with secondary growths of calcite) increases roughly linearly with time; this should be expected since the older the rock, the more the time in which it has been subjected to tectonic and other stresses. Open fissure density increases similarly to aggregate fissure density from recent through the mid-

Mesozoic; then, as calcite recrystallization is initiated, decreases the greater the age. Permeability, it is shown, is greatest the more recent the rock. This is due, according to Jakucs, not to structural fissuration, but to a high degree of textural porosity, and an unconsolidated, earthy consistency. After decreasing rapidly in the Cenozoic, as a result of compaction with time, it peaks again in the Mesozoic, presumably due to the increase in secondary permeability through fissuration overcoming the compaction due to time. These factors explain why the most conspicuous karst landscapes tend to occur most often in Triassic, Jurassic, and Cretaceous limestones.

Fig. 6.



Dense homogeneous limestones, which exhibit a conchoidal fracture, have been observed to dissolve much less rapidly than holocrystalline ones. This probably can be attributed to the small surface area of a smooth, conchoidal fracture as opposed to that of a jagged crystalline one. Not only does this textural characteristic lend itself to strong dissolution in a crystalline rock; but, since fracturing occurs along crystal faces (adhesion between crystals being weaker than the bonds holding together the crystal lattice), further surface area is exposed. During dissolution, minute etching pits will form, further increasing the area subject to dissolution (Jakucs 1977).

Bedding in limestone may involve 1) dividing surfaces, 2) a variation from the top of the bed to the bottom, or 3) lithologic differences between succeeding strata (Jennings 1971). Along bedding planes, insoluble constituents, such as clay and shale, are commonly present. These bedding planes inhibit karst development by allowing water passage only along these bedding planes. Although a high degree of bedding is widely considered unfavorable for karst; due to the abundance of insoluble residue, fissures may be filled-up, increasing the strength, and thereby at least partially negating the negative effect of bedding.

The surface slope of limestone is known to have an effect on the resultant landforms of a particular region. (Smith and Albritton 1941) describe three distinctive features as having resulted from the differences in slope. Their experiments were conducted in an arid region (Sierra Blanca, Texas), but the authors believe the

features to be dependent only upon the slope and; therefore, not specific to this particular area. The following features were observed on this jointed Cretaceous limestone:

1) Solution pits (tinajitas) : These are shallow, sub-circular depressions, forming only where water is allowed to pond, and on slopes $\leq 3^\circ$.

2) Solution facets : These are smooth surfaced blocks with a sharp-crested ridge on the upper portion due to water falling from joint gutters. These are formed on slopes of $3-6^\circ$. This is not sufficient slope to cause water to gather into rills.

3) Solution furrows : These are U-shaped troughs, having parallel or pinnate patterns. The divide between furrows is often reduced to a serrated ridge. These form on slopes great enough for water to collect into rills, concentrating its solvent action ($20^\circ- 90^\circ$).

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