

# **PS Evaluating the Relationship Between Stoichiometry and Cation Ordering in Ancient Dolomites\***

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Search and Discovery Article #51660 (2020)\*\*

Posted May 4, 2020

\*Adapted from poster presentation given at 2019 AAPG Annual Convention and Exhibition, San Antonio, Texas, May 19-22, 2019

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## **Abstract**

Over the past century, geologists have used various proxy resources to help explain the mechanisms responsible for dolomitization. Recently, laboratory experiments examining the fundamental controls on dolomite stoichiometry and cation ordering suggest that these mineralogical parameters may provide useful diagenetic information about the conditions of dolomitization. To test the applicability of these parameters to natural systems, this study presents a newly compiled global dataset of stoichiometry and cation ordering from over 1,200 dolomite samples from various geographic locations, geologic ages, and depositional and diagenetic environments. A number of relationships are examined, including the degree to which cation ordering and stoichiometry vary spatially and temporally, as well as the local and global geologic factors controlling the observed trends. The principal findings of this study are that: (i) dolomites formed in evaporitic settings are generally more stoichiometric than those formed in normal marine settings; (ii) dolomites associated with higher formation temperatures are generally more stoichiometric and well-ordered than their lower temperature counterparts; (iii) stoichiometry and cation ordering, though not co-dependent, may covary under certain dolomitization conditions; (iv) stoichiometric variations in stratigraphically related dolomites reflect dolomitization by evolving fluids. Taken together, these observations are inconsistent with the hypothesis that older dolomites are more stoichiometric because they have undergone a higher degree of recrystallization. Rather than reflecting global scale conditions, the data suggest that dolomite stoichiometry and cation ordering are controlled by local dolomitizing conditions, and thus make valuable proxies for understanding the conditions and fluids responsible for dolomitization.

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# Evaluating the Relationship between Stoichiometry and Cation Ordering in Ancient Dolomites

## Abstract

Over the past century, geologists have used various proxy resources to help explain the mechanisms responsible for dolomitization. Recently, laboratory experiments examining the fundamental controls on dolomite stoichiometry and cation ordering suggest that these mineralogical parameters may provide useful diagenetic information about the conditions of dolomitization. To test the applicability of these parameters to natural systems, this study presents a newly compiled global dataset of stoichiometry and cation ordering from over 1,200 dolomite samples from various geographic locations, geologic ages, and depositional and diagenetic environments. A number of relationships are examined, including the degree to which cation ordering and stoichiometry vary spatially and temporally, as well as the local and global geologic factors controlling the observed trends. The principal findings of this study are that: (i) dolomites formed in evaporitic settings are generally more stoichiometric than those formed in normal marine settings; (ii) dolomites associated with higher formation temperatures are generally more stoichiometric and well-ordered than their lower temperature counterparts; (iii) stoichiometry and cation ordering, though not co-dependent, may covary under certain dolomitization conditions; (iv) stoichiometric variations in stratigraphically related dolomites reflect dolomitization by evolving fluids. Taken together, these observations are inconsistent with the hypothesis that older dolomites are more stoichiometric because they've undergone a higher degree of recrystallization. Rather than reflecting global scale conditions, the data suggest that dolomite stoichiometry and cation ordering are controlled by local dolomitizing conditions, and thus make valuable proxies for understanding the conditions and fluids responsible for dolomitization.

## Problem, Objective & Hypothesis

### Problems:

- Few spatial and/or temporal databases containing dolomite stoichiometry and cation ordering exist.
- It has been nearly 40 years since the last major study of dolomite stoichiometry (Lumsden and Chimahusky, 1980) with no major studies focusing specifically on cation ordering.
- Numerous studies of ancient dolomites have reported dolomite stoichiometry and cation ordering. However, no comprehensive study has examined how these mineralogical parameters are related as well as how they vary based on age, depositional setting, physiographic setting.

### Objective:

- Here we have collected and analyzed 1,500+ dolomite samples to assess how these mineralogical parameters are related as well as how they vary based on age, depositional setting, physiographic setting.

### Hypothesis:

- It is posited that dolomite stoichiometry and cation ordering reflect localized physicochemical conditions.
- Further, it is suggested that dolomite stoichiometry and cation ordering may reflect secular variations in seawater chemistry through time and/or localized physicochemical conditions that are unique to various depositional and physiographic settings.

## Stoichiometry & Cation Ordering

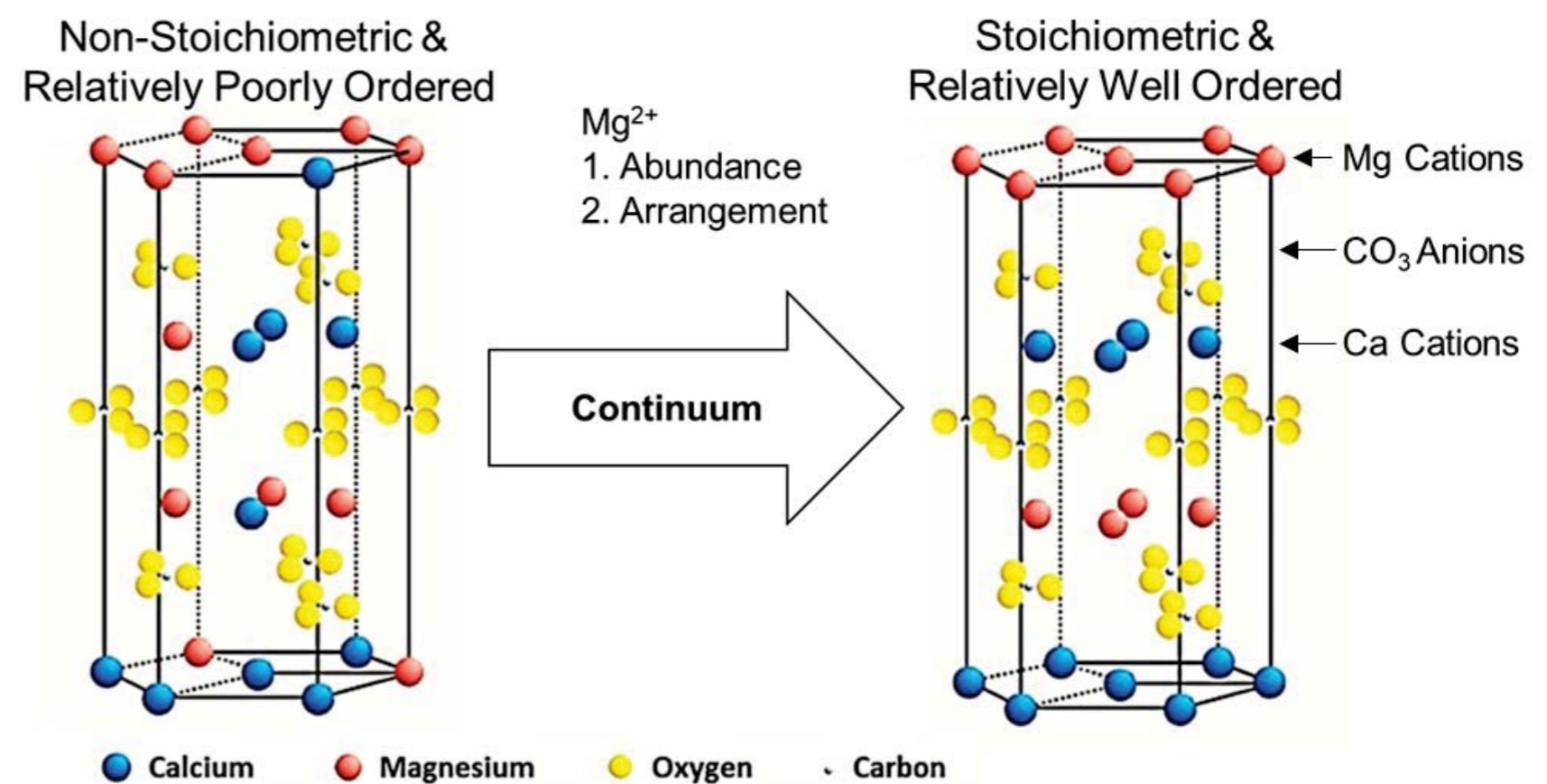


Fig. 1—Examples of non-stoichiometric relatively poorly-ordered dolomite and stoichiometric relatively well ordered dolomite crystal structures (Modified from Scholle and Ulmer-Scholle, 2003).

### Dolomite [CaMg (CO<sub>3</sub>)<sub>2</sub>] is defined by two mineralogical parameters:

Refer to Lippmann (1973), Reeder (1983), and Gregg et al., (2015).

**Stoichiometry (Composition):** The relative abundance of Mg to Ca in the crystal lattice (Fig. 1). Determined from the position of the D-(104) peak (Fig. 2) (Goldsmith and Graf, 1958). Dolomite stoichiometry is reported in various units (e.g., %Ca, mol% Ca, mole% CaCO<sub>3</sub>, mol% Mg, mole% MgCO<sub>3</sub>) all referring to the 1:1 molar ratio of CaCO<sub>3</sub>:MgCO<sub>3</sub>.

**Cation Ordering (Structure):** The degree to which the Mg and Ca cations are distributed into the appropriate cation layer within the crystal lattice (Fig. 1). Dolomite identified based on the presence of the D-(101), D-(015), and D-(021) peak (Fig. 2) (Goldsmith and Graf, 1958). The degree of cation ordering is reported as the ratio of D-(015):D-(110) peak height intensity (Goldsmith and Graf, 1958) or as hexagonal unit cell parameters (a ≈ 4.81 Å; c ≈ 16.05 Å) (Land, 1980).

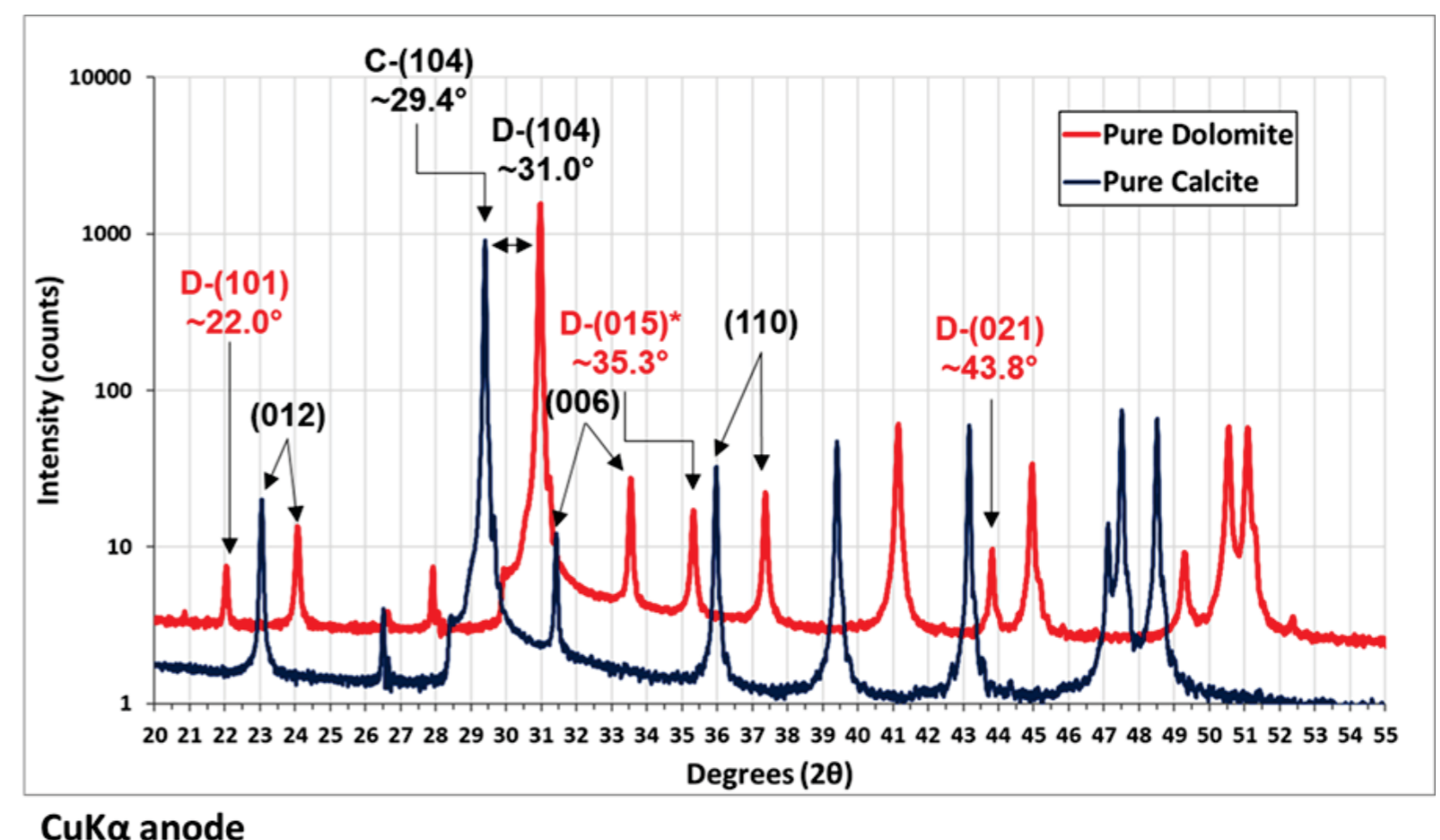


Fig. 2—Example X-ray diffractogram of mineralogically pure dolomite and calcite standards. Dominant peaks are labelled with dolomite ordering peaks labelled in red.

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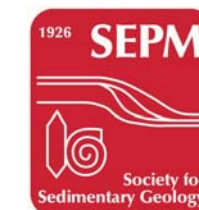
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## Global Dataset & Methodology



Location	Period (Epoch)	n	Depositional Setting	Physiographic Setting
Central Texas, U.S.A.	Cretaceous (Early)	346	Intertidal - Shallow	Platform Shelf
Utah, U.S.A.	Paleogene (Eocene)	149	Shallow	Lacustrine
Bonaire	Neogene (Miocene)	102	Shallow	Isolated Carbonate Platform
Michigan U.S.A.	Mississippian (Middle)	9	Shallow	Platform Shelf
	Devonian (Middle)	31	Shallow	Platform Shelf
	Devonian (Early)	119	Intertidal	Restricted Basin
	Silurian (Wenlock)	130	Shallow	Platform Shelf
	Silurian (Llandovery)	13	Shallow	Platform Shelf
	Ordovician (Middle)	2	Shallow	Platform Shelf
Curaçao	Neogene (Miocene)	6	Shallow	Isolated Carbonate Platform
Jamaica	Neogene (Pliocene)	1	Shallow	Isolated Carbonate Platform
Kitadaitōjima, Japan	Neogene (Pliocene)	6	Shallow	Isolated Carbonate Platform
South Florida, U.S.A.	Paleogene (Eocene)	11	Intertidal	Platform Shelf
Kentucky, U.S.A.	Mississippian (Middle)	5	Shallow	Platform Shelf
	Silurian (Wenlock)	1	Shallow	Platform Shelf
	Silurian (Llandovery)	1	Shallow	Platform Shelf
	Ordovician (Late)	2	Shallow	Platform Shelf
Southeast New Mexico/West Texas, U.S.A.	Permian (Guadalupian)	13	Shallow	Platform Shelf
	Permian (Cisuralian)	5	Shallow	Platform Shelf
	Ordovician (Early)	1	Shallow	Platform Shelf
Belize	Quaternary (Eocene)	4	Intertidal	Platform Shelf
Western Illinois	Mississippian (Late)	3	Shallow	Platform Shelf
	Mississippian (Middle)	41	Shallow	Platform Shelf
	Ordovician (Late)	2	Shallow	Platform Shelf
Northern Alberta, Canada	Devonian (Middle)	2	---	---
Southern Alabama, U.S.A.	Jurassic (Late)	16	Shallow	Platform Shelf
Northeastern Tennessee	Cambrian (Furongian)	1	Intertidal	Platform Shelf

Table 1—Samples collected for this study categorized by geographic location, period and epoch, deposition and physiographic setting.

Location	Period (Epoch)	n	Depositional Setting	Physiographic Setting	Study
Libya	Paleogene (Paleocene)	153	Shallow	Platform Shelf	Füchtbauer and Goldschmidt, 1965
Germany	Jurassic (Late)	91	Shallow	Platform Shelf	
Germany	Permian (Lopingian)	31	Intertidal	Restricted Basin	
Belgium	Devonian (Late)	3	---	---	
Abu Dhabi, U.A.E.	Quaternary (Holocene)	17	Intertidal	Platform Shelf	McKenzie, 1981
Yucatan Shelf	Cretaceous (Early)	4	---	---	
Levantine Basin	Neogene (Miocene)	1	---	---	
Continental Rise, W. Africa	Cretaceous (Late)	1	---	---	
Colombian Basin	Neogene (Pliocene)	1	---	---	
Cape Verde Basin	Cretaceous (Late)	1	Deep	Deep Basin	Lumsden, 1988
Tyrrhenian Basin	Neogene (Pliocene)	1	---	---	
Florida Escarpment	Cretaceous (Early-Late)	2	---	---	
Campeche Escarpment	Cretaceous (Early)	2	---	---	
Eastern Saudi Arabia	Quaternary (Pleistocene)	97	Intertidal	Platform Shelf	Chaffetz and Rush, 1994
Central Saudi Arabia	Jurassic (Late)	35	Shallow	Platform Shelf	
	Permian (Lopingian)	16	Intertidal	Platform Shelf	Banat et al., 1997
Northeastern S. Korea	Ordovician (Middle)	18	Intertidal	Platform Shelf	Chan Min Yoo and Yong Il Lee, 1998
Belgica Carbonate Mound Province, Porcupine Seabight	Neogene (Miocene)	4	Deep	Deep Basin	Gregg and Frank, 2009
Northeastern Bulgaria	Devonian (Middle)	14	Intertidal	Platform Shelf	Andreeva et al., 2011
Northern Italy	Triassic (Late)	28	Intertidal	Platform Shelf	Geske et al., 2012
Abu Dhabi, U.A.E.	Quaternary (Pleistocene)	2	Intertidal	Restricted Basin	
Austria	Triassic (Late)	1	Intertidal	Restricted Basin	
Belgium	Mississippian (Middle)	1	Intertidal	Restricted Basin	
Germany	Neogene (Miocene)	1	Shallow	Lacustrine	
---	Jurassic (Late)	2	Shallow	Platform Shelf	
---	Triassic (Late)	2	Shallow	Lacustrine	
---	Pennsylvanian (Early)	1	Shallow	Lacustrine	
---	Devonian (Middle)	1	Shallow	Platform Shelf	Geske et al., 2015
Greece	Triassic (Late)	1	Intertidal	Platform Shelf	
---	Permian (Lopingian)	1	Intertidal	Platform Shelf	
Kansas, U.S.A.	Permian (Lopingian)	1	Intertidal	Restricted Basin	
New Mexico, U.S.A.	Permian (Guadalupian)	2	Intertidal	Restricted Basin	
Northern Italy	Triassic (Middle)	1	Shallow	Platform Shelf	
Barbados	Quaternary (Pleistocene)	1	Shallow	Isolated Carbonate Platform	Winkelstern et al., 2016
Northeastern Australia	Neogene (Miocene)	18	Shallow	Platform Shelf	Veillard et al., 2019
Qatar	Paleogene (Eocene)	50	Shallow	Platform Shelf	Ryan and Kaczmarek (in preparation)

Table 2—Samples collected from previous studies categorized by geographic location, period and epoch, deposition and physiographic setting.

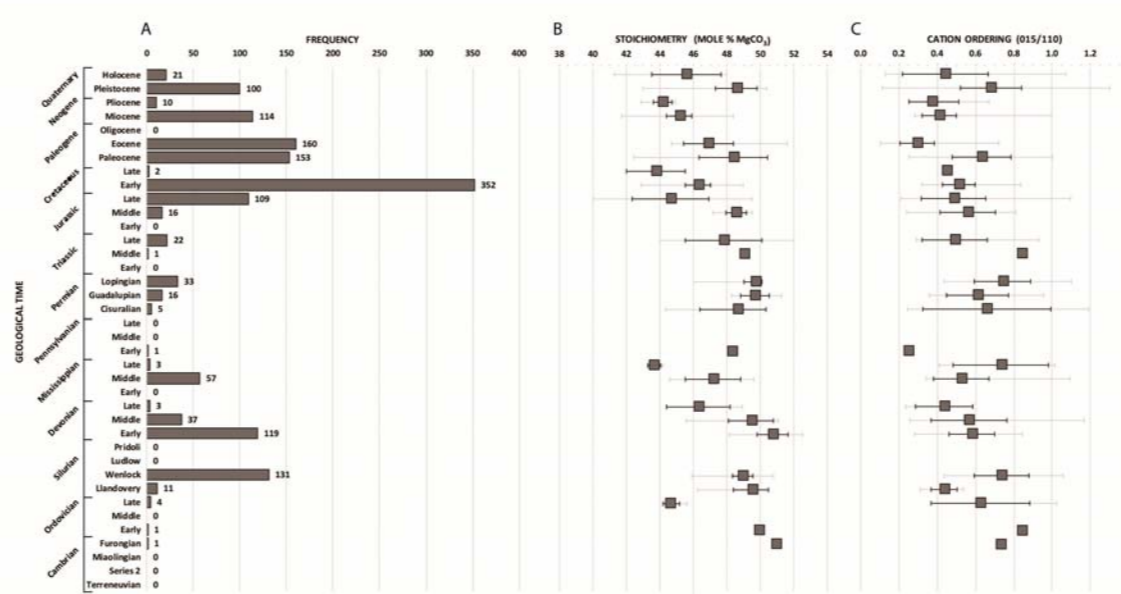


Fig. 4—(A) Frequency distribution of samples organized by epoch. (B) Mean stoichiometry per epoch. (C) Mean cation ordering per epoch. Black whiskers represent 1σ. Light gray whiskers represent minimum and maximum ranges.

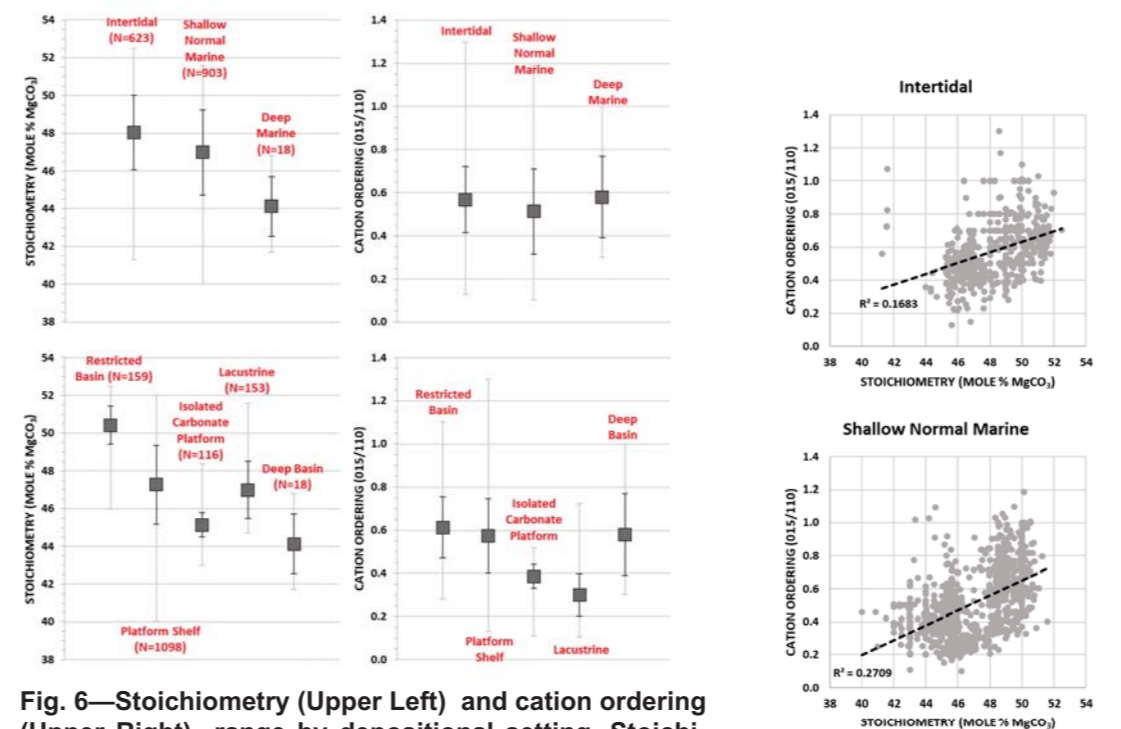


Fig. 6—Stoichiometry (Upper Left) and cation ordering (Upper Right) range by depositional setting. Stoichiometry (Lower Left) and cation ordering (Lower Right) range by physiographic setting.

**Key Observation:** Depositional settings with greater water depths tend to contain less stoichiometric dolomite.

## Results

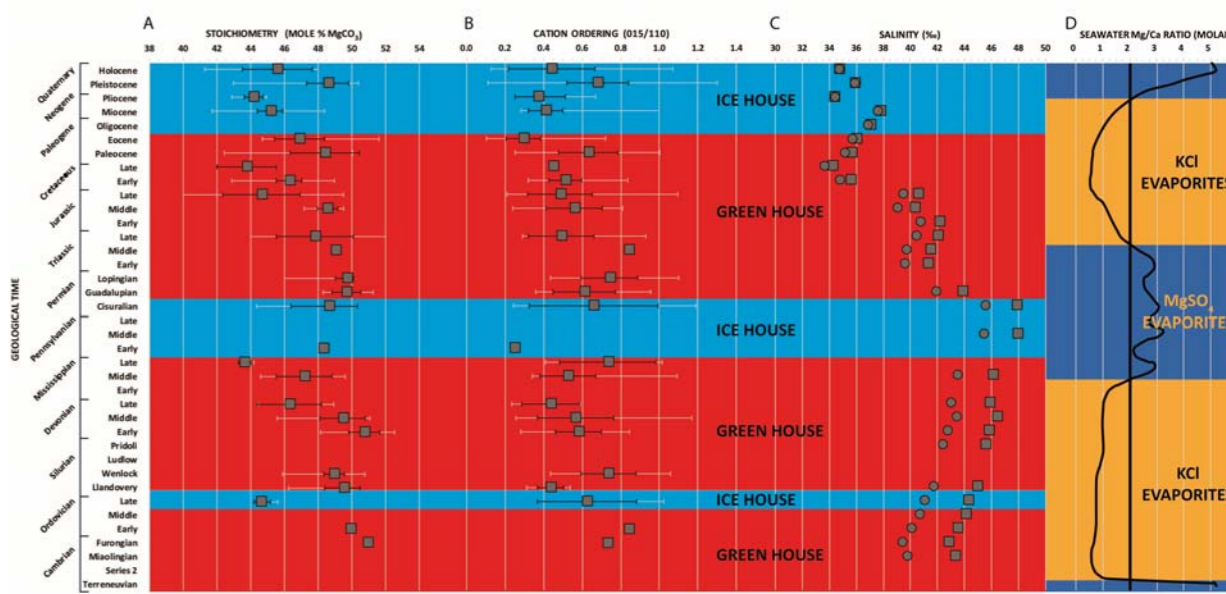


Fig. 5—(A) Mean stoichiometry per epoch. (B) Mean cation ordering per epoch. Black whiskers represent 1σ. Light gray whiskers represent minimum and maximum ranges. (C) Salinity models modified from Hay et al., (2006). (D) Seawater molar Mg/Ca ratio from Hardie (1996); the bold line divides the calcite and aragonite + High Mg Calcite nucleation field in seawater @25°C.

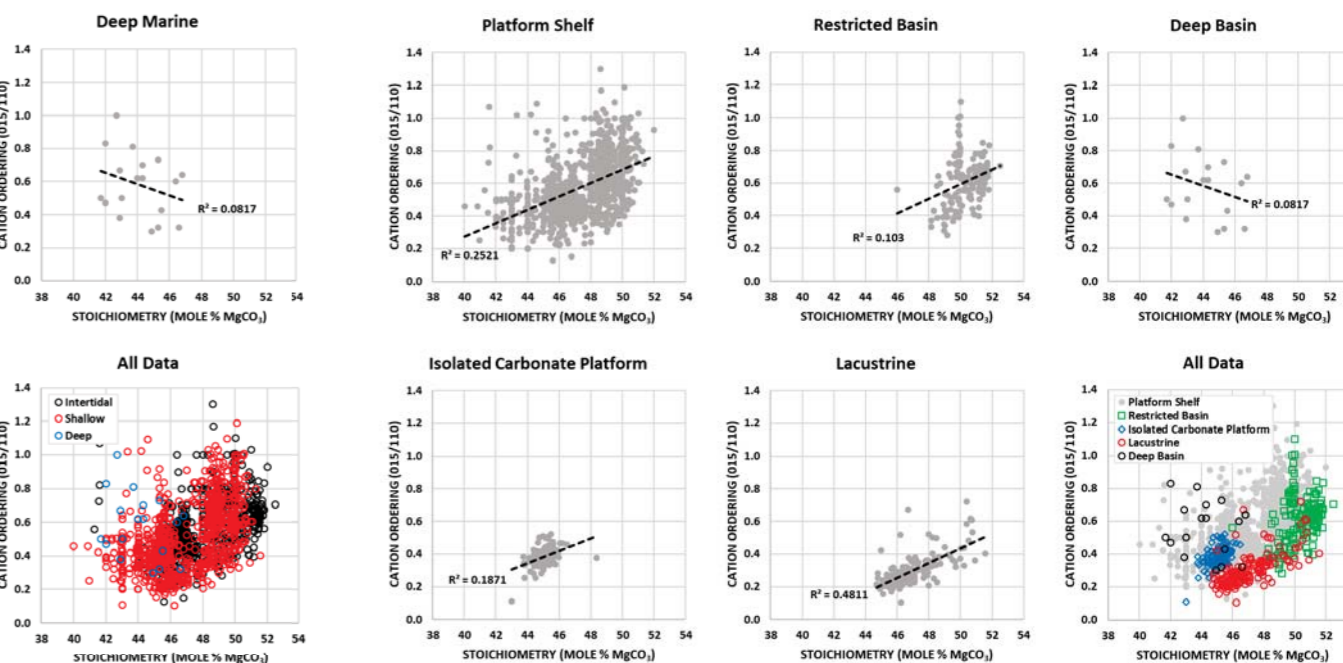


Fig. 7—Cross-plots of stoichiometry versus cation ordering based on depositional setting.

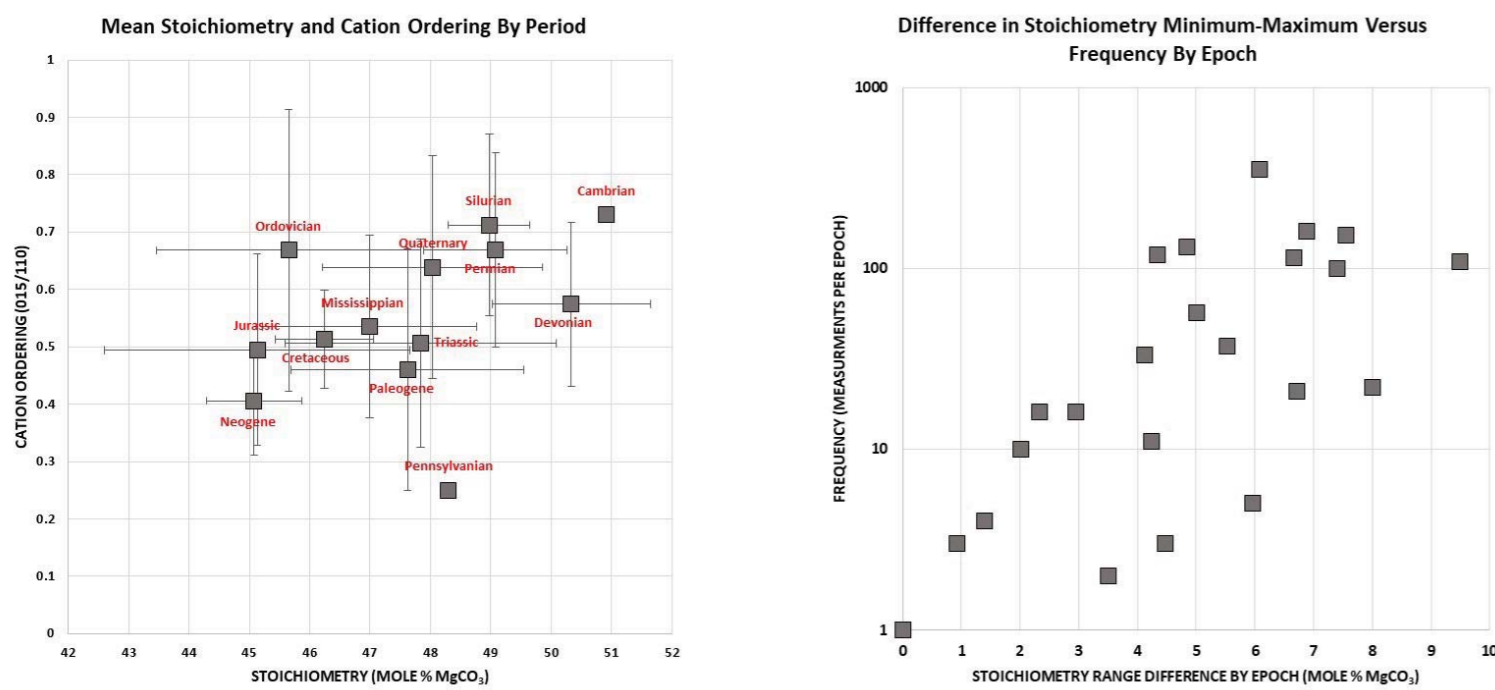
**Key Observation:** Stoichiometry and cation ordering do not statistically correlate.

Fig. 8—Cross-plots of stoichiometry versus cation ordering based on physiographic setting.

**Key Observation:** No statistical trends were observed based on physiographic setting, however, data from each physiographic setting tend to cluster with the exception of the platform shelf setting.

# Evaluating the Relationship between Stoichiometry and Cation Ordering in Ancient Dolomites

## Results Continued



**Fig. 9**—Cross-plot demonstrating the relationship between stoichiometry and cation ordering categorized by period (Left). Cross-plot demonstrating the relationship between the number of samples and the difference in stoichiometric range by epoch.

## Discussion & Limitations

- Dolomite stoichiometry and cation ordering appear to reflect localized physicochemical conditions rather than secular variations in seawater chemistry.
- The local physicochemical conditions of the dolomitizing fluid are suggested to reflect the depositional and/or physiographic setting.
- Work by Lumsden and Chimahusky (1980), Ren and Jones (2018), and Manche and Kaczmarek (2019) have demonstrated that stoichiometry may be useful proxy for understanding local changes in the dolomitizing fluid.
- However, this study is limited by two major factors:
  - (1) Complete information on samples provided by others or from literature. As a result, the depositional and physiographic setting have been overly simplified. For example, the platform shelf setting includes numerous sub-environments as such it shows significant variance in stoichiometry and cation ordering.
  - (2) Complete knowledge on whether samples have undergone recrystallization. Recrystallized dolomites exhibit stoichiometry composition and have high ordering. These samples will skew the dataset if included.

## Conclusions

- Statistical analysis of the number of samples versus the range in dolomite stoichiometry and cation ordering illustrate a bias in sample population. It is suggested that if more samples were added to the underrepresented ages that those ages would demonstrate equally large ranges. Thus, stoichiometry and cation ordering does not provide any predictive capabilities associated with age.
- Changes in dolomite stoichiometry and cation ordering do not correlate with secular variations in greenhouse/icehouse conditions, salinity, or seawater Mg/Ca ratios.
- Dolomite stoichiometry and cation ordering does not systematically increase with age refuting the notion of progressive recrystallization (Kupecz et al., 1993; Kupecz and Land, 1994).
- Local physicochemical conditions impact dolomite stoichiometry and cation ordering. Except for when dolomite is recrystallized.

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## Acknowledgements

The authors wish to thank Dr. David Budd, Dr. David Lumsden, Dr. Sal Mazzullo, Utah Geological Survey, and the Michigan Geological Repository for Research and Education for generously providing rock and powder samples. The authors also wish to thank Bill and Linda Harrison, Western Michigan University—Graduate College, and the Society for Sedimentary Geology for providing financial support to attend this conference.