

^{PS}Mg Isotopes in Hydrothermal Saddle Dolomites; Current Data for Paleozoic Dolomites of Eastern Canada and Implications for Mg Source*

Denis Lavoie¹, Simon Jackson², and Isabelle Girard²

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¹Geological Survey of Canada, Natural Resources Canada, Quebec City, QC, Canada (delavoie@nrcan.gc.ca)

²Geological Survey of Canada, Natural Resources Canada, Ottawa, ON, Canada.

Abstract

Hydrothermal dolomites are host to major hydrocarbon fields in North America. The process leading to the formation of these high temperature dolomites is controversial with end-members of 1) early, tectonically controlled, high temperature fluid migration upward along faults and laterally in porous limestones and 2) late, burial-dominated, regional migration of high temperature formation brines. In both cases, large amounts of magnesium are needed for the formation of the dolomite either as limestone replacement or void-filling cement. We present magnesium stable isotope ratios in saddle dolomites as a potential tool for recognition of Mg source and hence new data that could be of use in the ongoing debate.

In the Paleozoic of eastern Canada, saddle dolomites interpreted as hydrothermal are recognized in Ordovician shallow marine platform and slope carbonate, in Lower Silurian ramp carbonate and in Lower Devonian pinnacle reef. The occurrences are distributed from southern Quebec to the Gaspé Peninsula with the host successions occurring on lithologically diverse basement. The saddle dolomite samples were chemically characterized (ICP-ES) and their $\delta^{26/24}\text{Mg}_{\text{NBS88a}}$, $\delta^{25/24}\text{Mg}_{\text{NBS88a}}$ and $\delta^{26/25}\text{Mg}_{\text{NBS88a}}$ ratios measured (MC-ICP-MS).

The saddle dolomites in Lower Silurian carbonates in northern Gaspé were assumed to be sourced, through active foreland faulting, from underlying Ordovician ultramafic sivers; these dolomites and the adjacent fault-controlled dolomite bodies of Lower Ordovician slope carbonates are characterized by negative $\delta^{26/24}\text{Mg}_{\text{NBS88a}}$, $\delta^{25/24}\text{Mg}_{\text{NBS88a}}$ and $\delta^{26/25}\text{Mg}_{\text{NBS88a}}$ ratios (lower than, or around, 0‰ for the three ratios). On the other hand, Middle Ordovician saddle dolomites in southern Quebec are characterized by positive $\delta^{26/24}\text{Mg}_{\text{NBS88a}}$, $\delta^{25/24}\text{Mg}_{\text{NBS88a}}$ and

$\delta^{26/25}\text{Mg}_{\text{NBS88a}}$ ratios (over +1, +0.4 and +0.5‰, respectively). It is noteworthy that saddle dolomite in Lower Devonian pinnacle reef in northern Gaspé are adjacent to the ultramafic slivers but are characterized by positive Mg isotope ratios. This agrees well with the recently published model of magmatic dolomitizing fluids for this case.

There is no significant correlation between the chemical composition of the matrix and the Mg isotope ratios; it is preliminarily proposed that the isotopic variations could reflect different sources of Mg for dolomitization, but further research is needed to better constraint the applicability of that new tool.

Mg isotopes and hydrothermal saddle dolomites; current data for Paleozoic dolomites of eastern Canada and implications for Mg source

Denis Lavoie
Geological Survey of Canada – Québec, 490 de la Couronne, Québec City, QC
delavoie@rncan.gc.ca

Simon Jackson
Geological Survey of Canada – Ottawa, 601 Booth Street, Ottawa, ON

Isabelle Girard
Geological Survey of Canada – Ottawa, 601 Booth Street, Ottawa, ON

Introduction
Hydrothermal dolomites are host to major hydrocarbon fields in North America and abroad. The process leading to the formation of these high temperature dolomites is controversial and end-members that consist of 1) early, tectonically controlled, high temperature fluid migration upward along faults and laterally in porous limestones and 2) late, burial-dominated, regional migration of high temperature formation brines. In both cases, large amounts of magnesium (Mg) are needed for the formation of the dolomite, either as limestone replacement or void-filling cement. We present magnesium stable isotope ratios in saddle dolomites as a potential tool for recognition of Mg source and hence new data that could be of use in the ongoing debate.

Material and Methods

In the Paleozoic of eastern Canada, saddle dolomites interpreted as hydrothermal in origin are recognized in Lower Ordovician shallow marine platform and carbonate in Québec and Newfoundland, in Middle to Upper Ordovician carbonate basins such as the Trenton-Black River (TBR) interval, in the Lower Silurian ramp carbonate and in Lower Devonian pinnacles reef both in northern Gaspé Peninsula. The various occurrences of saddle dolomites that have been analyzed are distributed from southern Québec to the Gaspé Peninsula with the host successions overlying tectonically dissimilar basements with the potential of having Mg derived from diverse sources. The saddle dolomite samples were chemically characterized (ICP-ES) and their $\delta^{26}\text{Mg}_{\text{SMOW}}$, $\delta^{27}\text{Mg}_{\text{SMOW}}$ and $\delta^{28}\text{Mg}_{\text{SMOW}}$ ratios measured (MC-ICP-MS).

Results

The saddle dolomites in Lower Silurian carbonates in northern Gaspé are related to fluid circulation along the Shickook Sud Fault. Their Mg was hypothesized to have been sourced, through active brecciated faulting, from underlying Ordovician ultramafic dikes (Lavoie and Morin, 2004). The saddle dolomite precipitated from a saline (21–28 wt% NaCl_{eq}), high temperature fluid (150 to 200°C) with very positive $\delta^{26}\text{Mg}_{\text{SMOW}}$ values (+8 to +10) (Lavoie and Chi, 2010). In northern Gaspé, one field occurrence of Lower Ordovician saline carbonates was found associated with the Rivière Madeleine Sulf. However, temperature data are not available from these saddle dolomites. These first two intervals with saddle dolomites are characterized by $\delta^{26}\text{Mg}_{\text{SMOW}}$, $\delta^{27}\text{Mg}_{\text{SMOW}}$ and $\delta^{28}\text{Mg}_{\text{SMOW}}$ values lower than or around 0‰.

Middle Ordovician (or TBR) saddle dolomites are found in the St. Lawrence Platform and on Anticosti Island (Lavoie et al., 2005; Lavoie and Chi, 2010). They are, in the field and subsurface, associated with various faults that connected with the underlying Precambrian basement. The saddle dolomites precipitated out of a saline (20–25 wt% NaCl_{eq}), high temperature fluid (90 to 120°C) with positive $\delta^{26}\text{Mg}_{\text{SMOW}}$ values (+3 to +6) (Lavoie and Chi, 2010). These saddle dolomites are characterized by $\delta^{26}\text{Mg}_{\text{SMOW}}$, $\delta^{27}\text{Mg}_{\text{SMOW}}$ and $\delta^{28}\text{Mg}_{\text{SMOW}}$ values over +1, +0.4 and +0.5‰, respectively.

A massively dolomitized Lower Devonian pinnacle reef in northern Gaspé (Lavoie et al., 2010) is found at the junction between the Shickook Sud and Rivière Madeleine Sulf faults. The dolomitized body occurs 50 km SSW of the Lower Ordovician dolomite and 15 km to the E an occurrence of Lower Silurian dolomite. The massive replacement saddle dolomite formed from a saline (9 to 17 wt% NaCl_{eq}), very high temperature fluid (200 to 250°C) with elevated $\delta^{26}\text{Mg}_{\text{SMOW}}$ values (+7 to +10‰; Lavoie et al., 2010). Even though the dolomites are adjacent to the ultramafic dikes, they are characterized by positive Mg isotope ratios that strongly contrasted with those of the nearby Lower Ordovician and Lower Silurian dolomites of northern Gaspé. This specific massive replacement dolomite has been interpreted to originate from magmatic fluid and not from basinal brines that interacted with the ultramafic dikes in the adjacent basins (Lavoie et al., 2010). The new Mg stable isotope data indicates a different source of Mg compared with the nearby Lower Ordovician and Lower Silurian dolomites even though elements other than source alone could have involved in the Mg isotope ratios.

There is no significant correlation between the chemical composition of the matrix and the Mg isotope ratios, suggesting that the measured Mg isotope variations are not a product of instrumental matrix effects. It is, therefore, tentatively proposed that the isotopic variations reflect different sources of Mg for dolomitization, although conditions of dolomitization (e.g., temperature, rate of dolomite growth, precipitation versus replacement phases) may also have controlled isotopic fractionation between the dolomite itself and the dolomite host and have influenced the measured values. As the time of writing of this abstract, procedures are being developed for chemical separation of Mg from complex matrices. This will allow isotope ratios of potential Mg sources for dolomitization in the Gaspé (Lower Ordovician ultramafic dikes and shales) and southern Québec (Upper Ordovician shales) to be determined. Results should be available for discussion. The time scale is ± 10 Ma.

Conclusions

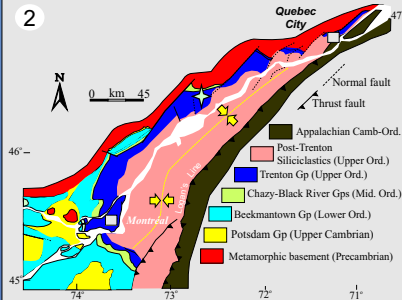
The current data suggest that Mg isotopes can offer important insights into sources and processes involved in dolomitization but further research is needed to better constrain the applicability of this new tool.

References

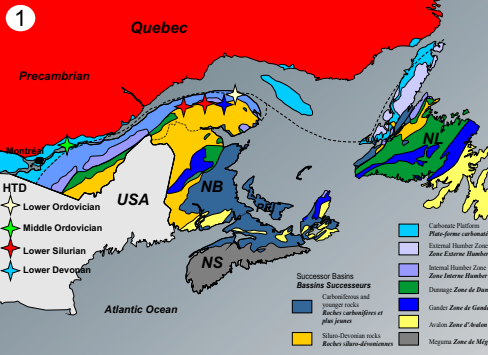
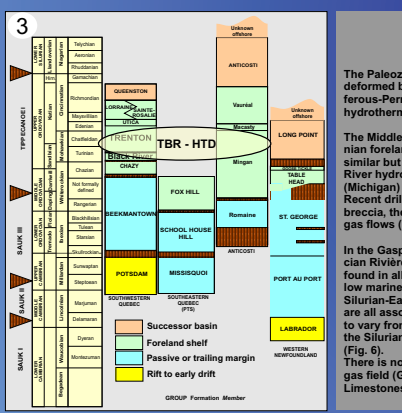
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IN A HURRY? - CONCLUSIONS

The ^{26}Mg and ^{25}Mg ratios of Lower Paleozoic saddle dolomites of interpreted hydrothermal origin have been analysed. This was done in an attempt to document the applicability of this geochemical tool to discriminate the diagenetic systems and source of Mg for dolomitization. The preliminary data confirm thermal fractionation of the Mg isotopes with higher temperature dolomites having more negative ^{26}Mg and ^{25}Mg ratios. Moreover, the geochemical character of the diagenetic fluids also play a significant role in the ^{26}Mg and ^{25}Mg composition of the dolomites as indicated by the relationships between the Mg isotopes and the $\delta^{87}\text{Sr}/\delta^{86}\text{Sr}$ and the $^{18}\text{O}_{\text{SMOW}}$ fluid ratios recorded by the dolomites. Current work focuses on other Paleozoic dolomites and the characterization of Mg source (ultramafic and shales).



Fracture-fill saddle dolomite in Trenton - Black River groups

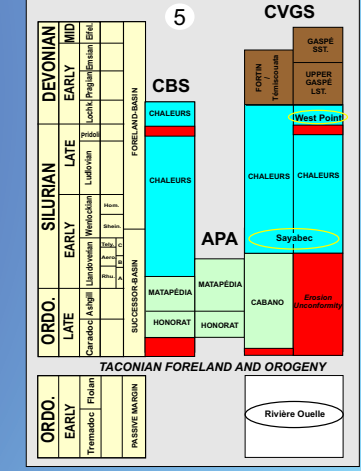
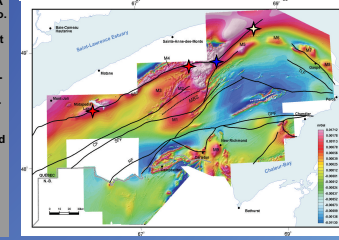
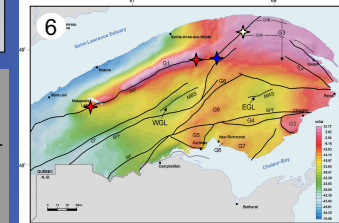
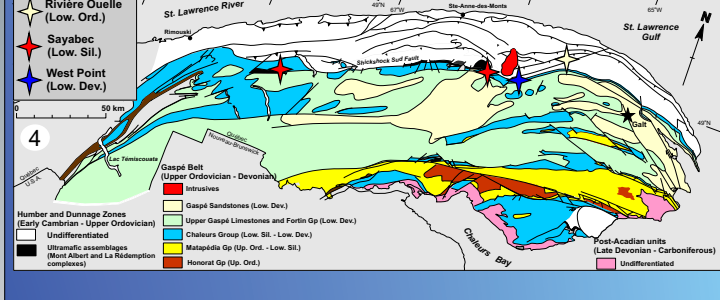


GEOLOGICAL, STRATIGRAPHIC AND TECTONIC SETTING

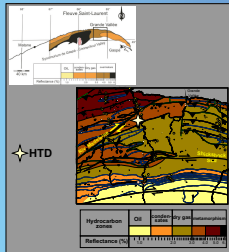
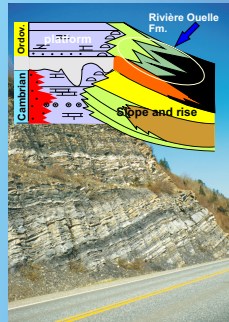
The Paleozoic successions of eastern Canada consists of 3 major domains (Fig. 1): The Cambrian-Ordovician deformed by the Taconian Orogeny, the Silurian-Devonian deformed by the Acadian Orogeny and the Carboniferous-Permian deformed by the Alleghanian Orogeny. This poster presents field and geochemical data on hydrothermal dolomites found in the first two geological domains of eastern Canada (Fig. 1).

The Middle to Late Ordovician Trenton-Black River of southern Québec (Fig. 2) represents the youngest Taconian foreland basin carbonates. Equivalent units are found in the Anticosti basin (Mingan; Fig. 3) whereas similar but older facies are present in western Newfoundland (Table Head Group; Fig. 3). The Trenton-Black River hydrothermal dolomites are major hydrocarbon reservoirs in the various intracratonic basins in the USA (Michigan) but also in the Appalachian basin of New York and in the St. Lawrence platform of southern Ontario. Recent drilling in the St. Lawrence platform in southern Quebec has confirmed the presence of dolomite-breccia, the distribution of which is controlled by faults. These dolomite breccia have yielded some significant gas flows (up to 9 mcmcf/d).

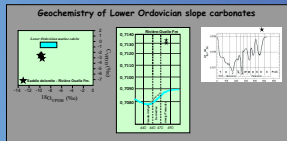
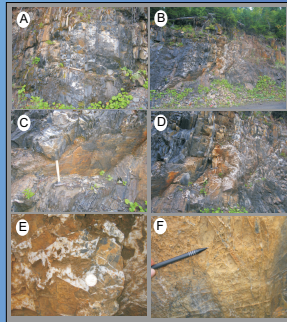
In the Gaspé Peninsula of eastern Québec, interpreted hydrothermal dolomites are found in the Lower Ordovician Rivière-Ouelle Formation of the Taconian fold and thrust belt (Fig. 4). Hydrothermal dolomites are also found in all major shallow marine carbonate units of the Silurian-Devonian Gaspé Belt, these include the shallow marine carbonate ramp of the Early Silurian Sayabec Formation and the major reef platform of the Late Silurian-Early Devonian West Point Formation (Figs. 4 and 5). These dolomite bodies in the Gaspé Peninsula are all associated with major Acadian dextral strike-slip faults although their mode of formation is documented to vary from one case to the other. It is noteworthy that the known occurrences of hydrothermal dolomites in the Silurian-Devonian Gaspé Belt are closely associated with both magnetic and gravity highs in the basin (Fig. 6). There is no current hydrocarbon production from the Sayabec and West Point formations, however, a small gas field (Galt Field) is hosted in a fault-controlled dolomitic breccia in the Lower Devonian Upper Gaspé Limestones in eastern Gaspé (Figs. 4 and 5).



LOWER ORDOVICIAN DOLOMITE

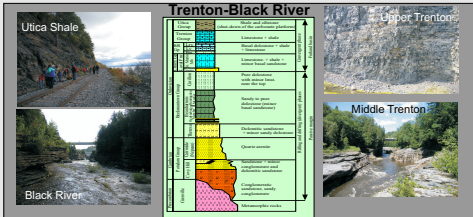


(lower left). The outcrop is cut by transtensional faults (top right, B and C). Massive dolomitization is controlled by the fault (top right D) with massively dolomitized fault breccia (top right E) and boxwork texture (top right F). The saddle dolomite cement in breccia is characterized by negative ^{18}O and ^{13}C ratios (lower right). However, the saddle dolomite cement is characterized by anomalously high strontium isotope ratios (lower right) which indicate that the dolomitizing fluid had intense interactions with the abundant underlying feldspar-rich siliciclastic units. No direct measurements of fluid temperatures are currently available.

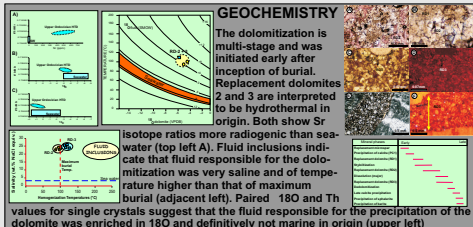
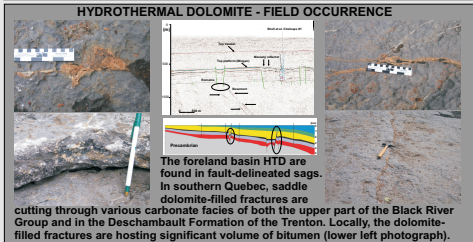


The Rivière Ouelle Formation is a Lower Ordovician slope and rise succession deposited during the passive margin stage of Laurentia (top left); it is a mixed carbonate and siliciclastic unit. In northern Gaspé Peninsula (eastern Canada), the Rivière Ouelle Formation occurs in the Taconian fold and thrust Belt. Near the trace of a major dextral strike-slip fault, an outcrop of massive dolomite interpreted to be of hydrothermal origin is found

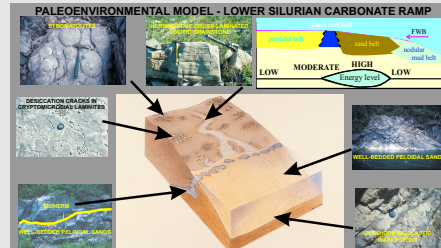
UPPER ORDOVICIAN DOLOMITE



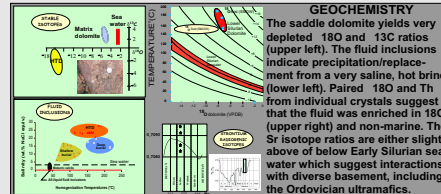
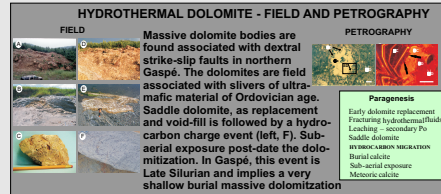
The Middle-Upper Ordovician Black River and Trenton groups are host to world-class hydrocarbon reservoirs in hydrothermal dolomites. They both represent shallow-marine carbonate units deposited in the Taconian foreland basin.



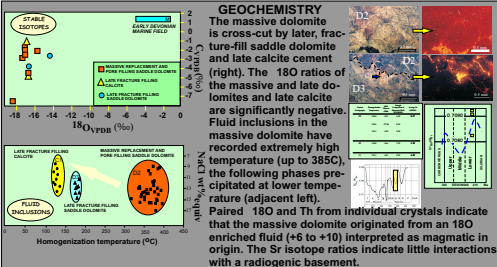
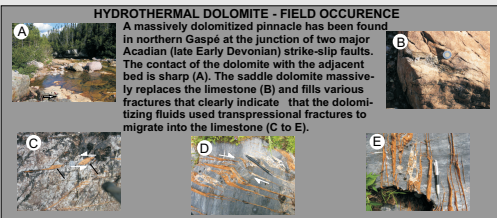
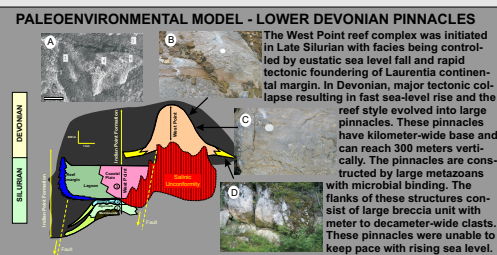
LOWER SILURIAN DOLOMITE

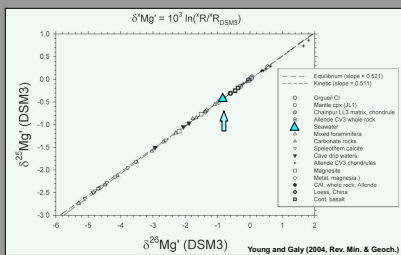


The Lower Silurian facies were deposited on a wide peritidal carbonate ramp at the onset of the Acadian foreland basin. The facies include a diversified peritidal assemblage of cryptomicrobial features; this belt is limited, seaward, by a narrow zone with metazoan-microbial bioherms followed by a zone with well bedded carbonate sands and ultimately by offshore muds. The facies evolution is controlled by eustatic sea level fluctuations and local tectonic collapse.



LOWER DEVONIAN DOLOMITE





Mg isotopes reservoirs

Magnesium has three stable isotopes: ^{24}Mg (78.99%), ^{25}Mg (10%) and ^{26}Mg (11.01%). Within natural Mg-rich elements, variations of the ^{26}Mg are less than 4‰. Seawater, because of its relatively long residence time has a fairly constant $\delta^{26}\text{Mg}$ of -0.82‰ (Adjacent left).

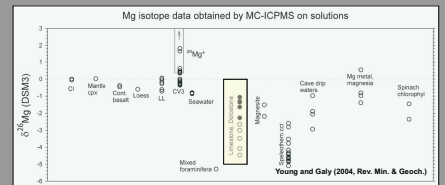
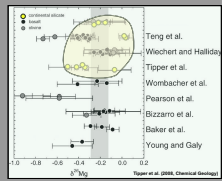
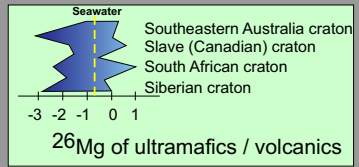
Ultramafics and mafic volcanic rocks are likely major source of Magnesium, the $\delta^{26}\text{Mg}$ signature of Precambrian and Phanerozoic ultramafic and mafic rocks is quite variable (-3 to +1‰) and averages at around -1 to -1.5‰ (left, center).

Continental-derived sediments, including shales, can also be considered as a potential source of Magnesium in diagenetic processes. There is a wide variability of $\delta^{26}\text{Mg}$ ratios in siliciclastic Mg-bearing mineral phases but, commonly, these are less negative than the oceanic (mafic) components (lower left).

Carbonates (both limestone and dolostone) preferentially incorporate light Mg isotopes, dolomite being more prone to heavier Mg isotope incorporation (see below right).

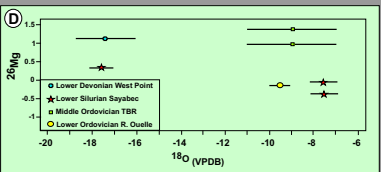
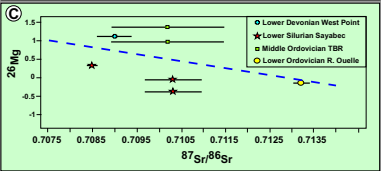
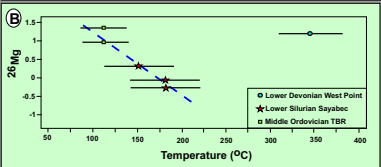
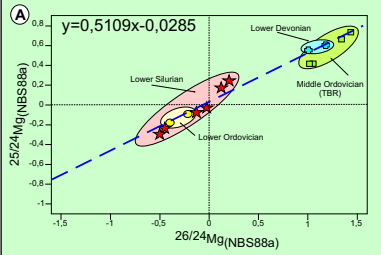
Finally, in natural three isotope systems (^{252}Mg and ^{250}Mg), fractionation in isotopic equilibrium will result in a regression line with a slope of 0.521 whereas if thermal or kinetic fractionation is involved, then the line will have a slope of 0.511.

This study of Mg isotopes aims at documenting, if any, variations in the isotope ratios of saddle dolomite.

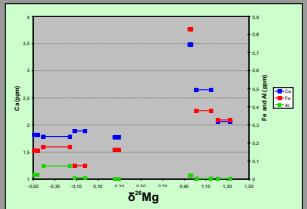


CONCLUSIONS

The ^{26}Mg and ^{25}Mg ratios of Lower Paleozoic saddle dolomites of interpreted hydrothermal origin have been analysed. This was done in an attempt to document the applicability of this geochemical tracer to characterize the diagenetic systems and source of Mg for dolomitization. The preliminary data confirm some thermal fractionation of the Mg isotopes with higher temperature dolomites having more negative ^{26}Mg and ^{25}Mg ratios. Moreover, the geochemical characteristics of the diagenetic fluids also play a significant role in the ^{26}Mg and ^{25}Mg composition of the dolomites as indicated by the relationships between the Mg isotopes and the $^{87}\text{Sr}/^{86}\text{Sr}$ and the $^{18}\text{O}_{\text{SMOW-FLUID}}$ ratios recorded by the dolomites. Current work focuses on other Paleozoic dolomites and the characterization of Mg source (ultramafics and shales).



| | Mg | Solution | Ca | Solution | Fe | Solution | Al | Solution | Mg | 2se | Mg | 2sd | nbs 2sd | Mg | 2se | Mg | 2sd | nbs 2sd |
|----------------|-------|----------|-------|----------|-------|----------|-------|----------|-------|------|------|------|---------|------|------|------|-----|---------|
| | ppm | | ppm | | ppm | | ppm | | % | % | % | % | % | % | % | % | % | % |
| Trenton | 1,020 | | 3,485 | | 0,830 | | 0,020 | | 0,96 | 0,07 | 0,02 | 0,34 | 0,40 | 0,04 | 0,00 | 0,20 | | |
| Trenton | 1,020 | | 3,485 | | 0,830 | | 0,020 | | 0,95 | 0,05 | 0,02 | 0,34 | 0,40 | 0,03 | 0,00 | 0,20 | | |
| BlackRiver | 1,021 | | 2,057 | | 0,327 | | 0,003 | | 1,21 | 0,08 | 0,16 | 0,58 | 0,61 | 0,04 | 0,05 | 0,29 | | |
| BlackRiver | 1,021 | | 2,057 | | 0,327 | | 0,003 | | 1,32 | 0,06 | 0,16 | 0,58 | 0,65 | 0,03 | 0,05 | 0,29 | | |
| Sayabec | 1,028 | | 1,819 | | 0,159 | | 0,025 | | -0,49 | 0,04 | 0,05 | 0,36 | -0,28 | 0,02 | 0,03 | 0,19 | | |
| Sayabec | 1,028 | | 1,819 | | 0,159 | | 0,025 | | -0,46 | 0,04 | 0,05 | 0,36 | -0,26 | 0,02 | 0,03 | 0,19 | | |
| Sayabec | 1,012 | | 1,889 | | 0,074 | | 0,007 | | -0,02 | 0,02 | 0,14 | 0,12 | -0,03 | 0,01 | 0,07 | 0,06 | | |
| Sayabec | 1,012 | | 1,889 | | 0,074 | | 0,007 | | -0,12 | 0,02 | 0,14 | 0,12 | -0,08 | 0,02 | 0,07 | 0,06 | | |
| Sayabec | 1,022 | | 1,772 | | 0,163 | | 0,001 | | 0,25 | 0,03 | 0,05 | 0,05 | 0,14 | 0,02 | 0,01 | 0,02 | | |
| Sayabec | 1,022 | | 1,772 | | 0,163 | | 0,001 | | 0,29 | 0,03 | 0,05 | 0,05 | 0,15 | 0,02 | 0,01 | 0,02 | | |
| West Point | 1,028 | | 2,650 | | 0,378 | | 0,004 | | 1,15 | 0,02 | 0,19 | 0,49 | 0,56 | 0,02 | 0,08 | 0,23 | | |
| West Point | 1,028 | | 2,650 | | 0,378 | | 0,004 | | 1,01 | 0,04 | 0,19 | 0,49 | 0,51 | 0,02 | 0,08 | 0,23 | | |
| Rivière Ouelle | 1,021 | | 1,783 | | 0,178 | | 0,073 | | -0,41 | 0,05 | 0,35 | 0,23 | -0,21 | 0,02 | 0,15 | 0,11 | | |
| Rivière Ouelle | 1,021 | | 1,783 | | 0,178 | | 0,073 | | -0,16 | 0,06 | 0,35 | 0,23 | -0,10 | 0,04 | 0,15 | 0,11 | | |



Saddle dolomites from the Lower Ordovician Riviere Ouelle Formation, the Middle Ordovician Black River and Trenton groups, the Lower Silurian Sayabec Formation and the Lower Devonian West Point Formation were analyzed with ICP-MS for their ^{26}Mg and ^{25}Mg ratios together with major-minor elements content. The results are shown in the table above and the location of the samples is on the geological maps (first panel). The ^{26}Mg and ^{25}Mg ratios vary from -0.49 to 1.32 permil and from -0.28 to 0.65 permil, respectively. There is no relationship between the ^{26}Mg and ^{25}Mg ratios of dolomites and their Ca, Fe and Al content (upper right graph).

The plot of ^{26}Mg versus ^{25}Mg (left, A) shows a linear relationship with a correlation equation of $y=0,5109x-0,0285$. The correlation indicates that the fractionation of the magnesium isotope is controlled by kinematic processes. It is noteworthy that two major groupings are recognized: the first one consists of saddle dolomites with positive ^{26}Mg and ^{25}Mg ratios (Middle Ordovician TBR and the Lower Devonian West Point) and a second group made of the Lower Ordovician Riviere Ouelle and Lower Silurian Sayabec Formations with near 0 to negative ratios.

Discarding the very high temperature saddle dolomite of the Lower Devonian West Point Formation, a strong relationship exists between ^{26}Mg and temperature of precipitation (based on fluid inclusion Th) of saddle dolomite (left, B). Higher temperatures of precipitation are associated with negative ^{26}Mg ratios. Such relationship would explain the kinematic fractionation suggested by the ^{26}Mg and ^{25}Mg plot (A).

A less convincing relationship seems to link the ^{26}Mg and the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of saddle dolomite (left, C). However, this relationship (decreasing ^{26}Mg ratios with increasing radiogenic values) is only valid if the data point from the Riviere Ouelle Formation is valid. Such relationship would link the magnesium isotopic ratios of void-filling and replacement saddle dolomite with the nature of the dolomitizing fluid. Finally, if no clear relationship is detectable between ^{26}Mg and ^{18}O of saddle dolomite (left, D), a relationship is developed between the ^{26}Mg ratio of saddle dolomite and the $^{18}\text{O}_{\text{fluid}}$ data set is very limited (lower, E).

SUMMARY OF GEOCHEMICAL DATA

| Age - Unit | Fluid Inclusions (Th-°C) | $^{87}\text{Sr}/^{86}\text{Sr}$ | $^{18}\text{O}_{\text{VPDB-Dol}}$ (‰) | $^{18}\text{O}_{\text{VSMOW-Fluid}}$ (‰) |
|-----------------------------------|--------------------------|---------------------------------|---------------------------------------|--|
| Lower Ordovician - Riviere Ouelle | | 0.7131 - 0.7132 | -10 to -9 | |
| Middle Ordovician - TBR | 80 - 130 | 0.7088 - 0.7115 | -11 to -7 | +4 to +6 |
| Lower Silurian - Sayabec | | | | |
| - Ruisseau Isabelle section | 110 - 200 | 0.7082 - 0.7085 | -18 to -17 | |
| - Lac Matapedie section | 140 - 220 | 0.7096 - 0.7101 | -8 to -7 | +8 to +10 |
| Lower Devonian - West Point | 300 - 380 | 0.7084 - 0.7092 | -19 to -16 | +6 to +10 |