

Original Paper

Identifying the magnesium source in hydrothermal dolostone reservoirs: A multi-proxy approach using $\delta^{26}\text{Mg}$, $\delta^{44/40}\text{Ca}$, and $^{87}\text{Sr}/^{86}\text{Sr}$

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ARTICLE INFO

Article history:

Received 9 August 2025

Received in revised form

1 November 2025

Accepted 27 May 2026

Available online 3 June 2026

Edited by Min Li

Keywords:

Ediacaran

Yangtze Block

Saddle dolomite

Diagenetic model

Dolomitisation fluid

ABSTRACT

Dolostone, especially replacement dolostone, is a significant component of Earth's carbonate rock archive. Hydrothermal dolostones, often associated with fault zones, play a crucial role in the dolostone record. However, the origin—particularly the sources of magnesium involved in their formation—remains unclear. This study focuses on two case examples of upper Ediacaran hydrothermal dolomite from the Yangtze Block, utilising a multi-proxy approach that examines $\delta^{26}\text{Mg}$, $\delta^{44/40}\text{Ca}$, $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, and elemental concentrations. In the first study site (i.e., the central Sichuan Basin in the upper Yangtze Block), saddle dolomites of Ediacaran dolostones were measured to have fluid inclusion homogenization temperatures up to 260 °C, which are significantly higher than those the strata experienced, and are characterised by low $\delta^{26}\text{Mg}$ values ($-2.23\text{‰} \pm 0.18\text{‰}$) and $\delta^{44/40}\text{Ca}$ values (0.81‰ and 1.10‰), along with high strontium (Sr) contents (103.9 ± 56.0 ppm) and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (0.7119 ± 0.0017). These geochemical data indicate that hydrothermal dolomites were precipitated from hydrothermal fluids after obtaining Mg from chlorite-bearing crustal lithologies. Hydrothermal dolomitisation likely occurred in the mixing of crust-derived fluids and burial brines. In contrast, at the second study site (i.e., Nanshanping outcrop at the middle Yangtze Block), the presence of ^{26}Mg -enriched ($\delta^{26}\text{Mg}$: -0.6‰ to -0.5‰) saddle dolomite with low Sr contents (47.0 ± 9.6 ppm) suggests the influence of mantle-derived fluids. The hydrogeochemistry of the parent fluid appears to be the primary factor controlling the $\delta^{26}\text{Mg}$ and $\delta^{44/40}\text{Ca}$ values, as well as the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of hydrothermal dolomite. This is further verified by hydrothermal dolomite from the Ediacaran to the Phanerozoic and diagenetic models. Low $\delta^{26}\text{Mg}$ values combined with high $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in hydrothermal dolomite generally indicate the presence of crustal fluids. Conversely, high $\delta^{26}\text{Mg}$ values, along with low $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, are typically associated with the involvement of the mantle-derived fluids. The study presents two distinct sources of Mg, typically for the formation of hydrothermal dolostones, providing some clues to solve the puzzle for the origin of dolomite, particularly in settings characterised by widespread hydrothermal fluid activity.

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Peer review under the responsibility of China University of Petroleum (Beijing).

<https://doi.org/10.1016/j.petsci.2026.05.051>

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1. Introduction

Dolostones represent an important component of the global carbonate record (Immenhauser, 2022; Li et al., 2023; Lazar et al.,

2025). Primary marine or lacustrine dolomite (even in its non-stoichiometric form), in the sense of a direct precipitate from sea or lake water or porewater, is not common in recent surface environments (Petrash et al., 2017; Hood and Wallace, 2018; Hu et al., 2020a; Akhtar et al., 2024). The majority of dolomite minerals are formed by replacement dolomitisation of aragonite and calcite mineralogies and, if pervasive, form dolostone (the rock) across a broad spectrum of environments (see Prather et al. (2023) for a recent review). Of which, hydrothermal dolomitisation, often associated with fault zones, is replacement of calcium carbonate by dolomite under temperatures of 5 to 10 °C hotter than the surrounding host rocks (Fig. 1, Machel and Lonnee (2002)). This process is one of the most common processes responsible for variation in petrophysical properties in hydrocarbon reservoirs (Jiang et al., 2018; Hu et al., 2023a) and contributing to the formation of ore deposits (Giorno et al., 2022; Zhou et al., 2022).

Magnesium, as a key elemental constituent in dolomite formation, plays a pivotal role in the dolomitisation, which is intrinsically linked to the “dolomite problem” (Qing et al., 2023). Understanding the provenance of magnesium is fundamental to distinguishing different types of dolomites. Isotopic systems, particularly those of magnesium ($\delta^{26}\text{Mg}$), calcium ($\delta^{44/40}\text{Ca}$), and strontium ($^{87}\text{Sr}/^{86}\text{Sr}$), have been increasingly applied as geochemical tracers to constrain the nature of dolomitising fluids and history (Riechermann et al., 2020; Jiao et al., 2023; Muñoz-López et al., 2024). Temperature-fractionation functions for Mg isotope in ordered dolomite are applied to reconstruct palaeo-seawater properties (Li et al., 2015; Walter et al., 2015; Bialik et al., 2018; Higgins et al., 2018; Hu et al., 2019; Shalev et al., 2021; Zhang et al., 2022; Xia et al., 2024; Lazar et al., 2025). Compared to modern seawater (-0.83‰ , Shalev et al. (2019)),

mantle (-0.25‰ , Teng (2017)) and chlorite (-1.8‰ , Kimmig et al. (2021)) yield completely different $\delta^{26}\text{Mg}$ values. Correspondingly, a compilation of reported hydrothermal dolomite $\delta^{26}\text{Mg}$ values through Earth's history reveals either significantly higher or lower magnesium isotope values than that of the coeval host dolostone (Fig. 2(a)). Although several types of hydrothermal fluids have been proposed to explain the observed variability in $\delta^{26}\text{Mg}$ values (Azmy et al., 2013; Geske et al., 2015a; Walter et al., 2015; Kimmig et al., 2021; Araoka et al., 2022; Jiao et al., 2023; Muñoz-López et al., 2024), fingerprinting that the possibly multi-source origin of Mg-bearing fluids is still a challenge.

Due to variability and uncertainty in calcium isotopic fractionation, dolomite yields a wide range of fractionation factors depending on the fluid type involved and various kinetic factors (Fantle and Tipper, 2014; Blättler et al., 2015; Perez-Fernandez et al., 2017). This significantly complicates the application of calcium isotope as tracers for hydrothermal dolomitisation. Notably, limited data (Steuber and Buhl, 2006; Riechermann et al., 2020; Chen et al., 2023) have shown that fluids interacting with clastic crustal bedrock exhibit lower $\delta^{44/40}\text{Ca}$ values, whereas higher values are characteristic of fluids percolating through carbonate rocks. At elevated temperature, dolomite exhibits non-conservative Ca release during near-equilibrium dissolution without net mineral precipitation (Perez-Fernandez et al., 2017). Furthermore, Ca isotopic fractionation during calcite precipitation under hydrothermal conditions is considerably lower than that observed in low-temperature experiments and natural samples (Brown et al., 2013).

In addition, strontium isotope ratios ($^{87}\text{Sr}/^{86}\text{Sr}$) have been used to fingerprint magnesium sources of hydrothermal fluids (Fig. 2(b)–Qing and Mountjoy, 1994; Brown et al., 2013; Walter

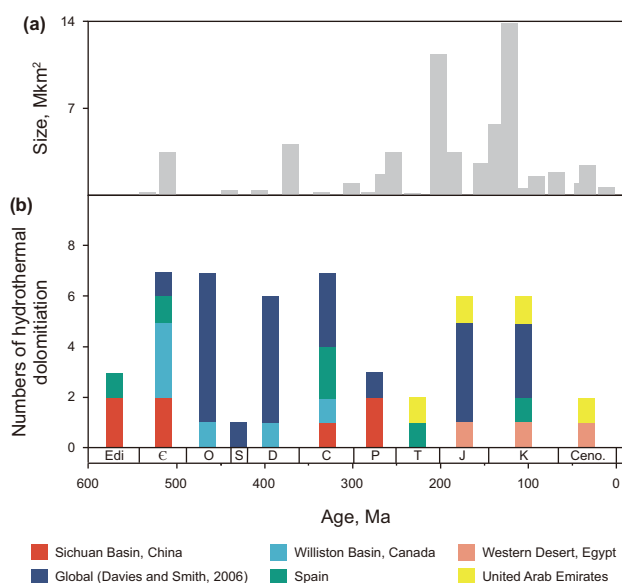


Fig. 1. Compilation of large Igneous Provinces (LIP) as well as hydrothermal dolomite cases in different basins for the Ediacaran and the Phanerozoic. (a) Compilation of LIP (Cather et al., 2009; Nechaev et al., 2022) for the Phanerozoic. (b) Count of hydrothermal dolomite cases in different basins over the Ediacaran and the Phanerozoic. Data from the Sichuan Basin are based on Liu et al. (2017), Zhao et al. (2018), Xiao et al. (2020), Liu et al. (2021a), Liang et al. (2022) and Hu et al. (2023a). Burial dolomite data from the Williston Basin are from Katz et al. (2006), Kimmig et al. (2021) and Muñoz-López et al. (2024). Burial dolomite data from the Western Desert (Egypt) are from Rifai et al. (2006) and Mazzini et al. (2019). Data from Spain are from Tritlla et al. (2001), Gasparrini et al. (2003, 2006), López-Horgue et al. (2010), Herrero et al. (2011) and Laponi et al. (2014). Data from the United Arab Emirates are based on Sirat et al. (2016), Alqattan (2022), Morad et al. (2023) and Mansurbeg et al. (2024).

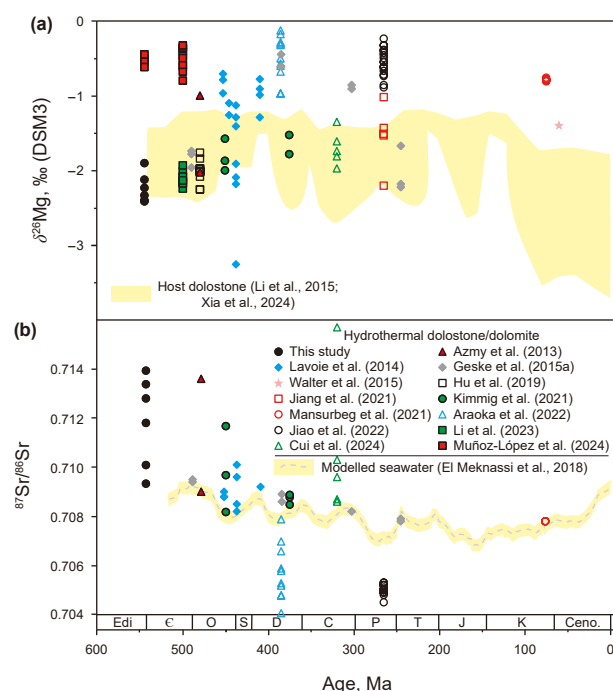


Fig. 2. The compilation of $\delta^{26}\text{Mg}$ (DSM3) values (a) and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (b) of hydrothermal/burial dolostone/dolomite, compared to host dolostone values (Li et al., 2015; Xia et al., 2024) and palaeo-seawater values (El Meknassi et al., 2018), from the Ediacaran to the Phanerozoic. Other data are from Azmy et al. (2013), Lavoie et al. (2014), Geske et al. (2015a), Walter et al. (2015), Hu et al. (2019), Jiang et al. (2021), Kimmig et al. (2021), Mansurbeg et al. (2021), Araoka et al. (2022), Jiao et al. (2023), Li et al. (2023), Cui et al. (2024) and Muñoz-López et al. (2024).

et al., 2015; Hu et al., 2023a). This is due to that mantle rocks are characterised by low $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (ca. 0.7035, Palmer and Elderfield, 1985; Salters and Stracke, 2004), while continental crust (sedimentary rocks are included), yield $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of 0.7400 (Wei and Algeo, 2020; Lenz et al., 2024).

Motivated by the complexity of replacement dolomitisation in the subsurface, we compare and discuss the widespread occurrence of hydrothermal dolostones in the upper Ediacaran Dengying Formation, Yangtze Block. Notably, the Dengying Formation represents a critical target for deep-burial and ultra-deep gas exploration in Sichuan Basin with proven gas reserve exceeding $600 \times 10^9 \text{ m}^3$ (Zhao et al., 2021). During the post-depositional stage, this formation in Yangtze Block underwent multiple extensional activities with complex hydrothermal alteration (Hu et al., 2023a; Muñoz-López et al., 2024). The upper Ediacaran hydrothermal dolostone from the Sichuan Basin, forming part of the upper Yangtze Block (Hu et al., 2023a), yield significantly different geochemical archive data compared to those of the Nanshanping outcrops, forming part of the middle Yangtze Block (Muñoz-López et al., 2024). However, the mixing of crustal fluids and burial brines are suggested to provide the magnesium source for hydrothermal dolomitisation in their studies. Here, we explore these two contrasting carbonate archives to decipher the magnesium source for hydrothermal dolomitisation by means of a multi-proxy $\delta^{26}\text{Mg}$, $\delta^{44/40}\text{Ca}$, $^{87}\text{Sr}/^{86}\text{Sr}$, and elemental concentrations approach combined with numerical modelling. Data are placed in context with published geochemical properties of hydrothermal dolomites from the Ediacaran to the Phanerozoic. This study aims to (i) compare magnesium sources in the upper Ediacaran hydrothermal dolomitisation of the Sichuan Basin and those of the Nanshanping outcrop based on $\delta^{26}\text{Mg}$ values, (ii) further provide insights into magnesium sources in the upper Ediacaran hydrothermal dolomitisation through $\delta^{44/40}\text{Ca}$ values and other geochemical proxy, and (iii) establish a general model of magnesium sources in hydrothermal dolomitisation across geological time.

2. Geological setting

The Sichuan Basin is an intracratonic basin and is located in the upper Yangtze Block, SW China (Fig. 3(a)). During the Neoproterozoic, the Yangtze area was subject to rifting and then evolved into a passive continental margin. The Neoproterozoic crustal clastic rocks are well-developed in the Sichuan Basin (He et al., 2017), while the Jiangnan Orogenic Belt, around the middle Yangtze Block, consists of mantle rocks due to a mantle superplume during the break up of the supercontinent Rodinia (Fig. 3(b); Li et al., 2003; Zhao and Cawood, 2012; Yao et al., 2019). In the late Ediacaran, an intracratonic rift developed within the Sichuan Basin (Hu et al., 2020a).

The upper Ediacaran Dengying Formation was deposited between 551.1 ± 0.7 and 541 Ma (Condon et al., 2005). In the Sichuan Basin, the Dengying Formation has a total stratigraphic thickness ranging from 200 to 900 m. It is divided into four members in ascending order (Fig. 3(c)). The first member is predominantly composed of dolostone and clastic rocks, and the second member consists of crystalline dolostone and microbialites. The third and fourth members comprise clastic rocks and dolostones, respectively. With respect to the sedimentary environments, the Dengying Formation in the Sichuan Basin consists of uplifted carbonate platforms, intra-platform basins and deep-water basins. The drilled cores and outcrops studied in the context of this paper are from the carbonate platform domain in the Sichuan Basin.

The upper Ediacaran Dengying Formation has undergone multiple phases of tectonic activity and hydrothermal overprint. The first stage of hydrothermal activity leads to the precipitation of

silica-occluded veins, chert, and cherty dolostone during the late Ediacaran to Cambrian (Chen et al., 2009; Hu et al., 2023a), while the second hydrothermal stage results in the presence of zebra dolostone, saddle dolomite, fluorite, and quartz and takes place between the Permian and the Jurassic (Hu et al., 2020b; Su et al., 2022). Moreover, the Permian extensional activity leads to the precipitation of large volumes of chlorite (12%–30%) in igneous rocks (Ma et al., 2019). The Dengying Formation reached a maximum burial depth of >8 km during the Cretaceous and was subsequently uplifted to the current burial depths of ca. 5 km (Hu et al., 2020b).

In the Nanshanping outcrop, the upper Ediacaran Dengying Formation was deposited in a shallow-water environment and is stratigraphically divided into three members (Cui et al., 2016). The basal Hamajing Member comprises dolomitised, stratiform stromatolites; the middle Shibantan Member consists of claystones, siltstones, and sandstones; and the upper Baimatuo Member is characterised by grey dolomitic stromatolites (Lu et al., 2024). During the post-depositional stage, compressional tectonic activities generated thrust faults, which served as fluid conduits transporting fluids to the Dengying Formation (Muñoz-López et al., 2024). Compared with the Sichuan Basin where the crustal thickness is approximately 42–46 km, the Nanshanping outcrop area is characterised by a thinner crust of about 32–34 km (Li et al., 2006).

3. Methodology

Hydrothermal dolostone and dolomite samples were initially collected from the second and fourth members of Dengying Formation from cores of 15 drilled wells and 4 outcrops across the Sichuan Basin for petrological observations, fluid inclusion homogenization temperatures, $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ analyses, and $^{87}\text{Sr}/^{86}\text{Sr}$ measurements, as reported in Hu et al. (2023a).

After further petrographic screening of all the thin sections of the above samples and investigating the geochemical data, representative, hydrothermal or saddle dolomite samples from 4 outcrops in the west and wells MX9, MX108, MX117, and MX118 in the central Sichuan Basin were selected for detailed elemental concentration, $\delta^{26}\text{Mg}$, and $\delta^{44/40}\text{Ca}$ analyses in this study (Table 2; Fig. 3). These samples were chosen to cover a broad range of $\delta^{13}\text{C}$ values (−3.0‰ to 0.5‰; Hu et al., 2023a) and to represent the isotopic variability of hydrothermal dolomite within the studied interval. These $\delta^{26}\text{Mg}$, and $\delta^{44/40}\text{Ca}$ analyses were conducted in the Laboratory of Sediment and Isotope Geology, Ruhr-University Bochum, Germany.

3.1. Petrography

Thin sections, 30 μm thick, were performed for petrographic observation. Furthermore, about 20 pieces of thin sections were stained with Alizarin Red S to differentiate calcite from dolomite. Cathodoluminescence analysis was conducted using a Relion III cold cathode microscope, operating with a 4 mm beam diameter, a current of 325 μA , and a beam voltage of 16.3 kV.

3.2. Geochemical analyses

3.2.1. Elemental concentrations

To analyse major and trace elemental contents, 1.5 mg of dolomite powder was leached in 1 mL 3 M HNO_3 for 24 h 2 mL H_2O was then added to the aliquots, which were finally analysed by Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES). The standards BCS-CRM512 (dolomite) and BCS-CRM513

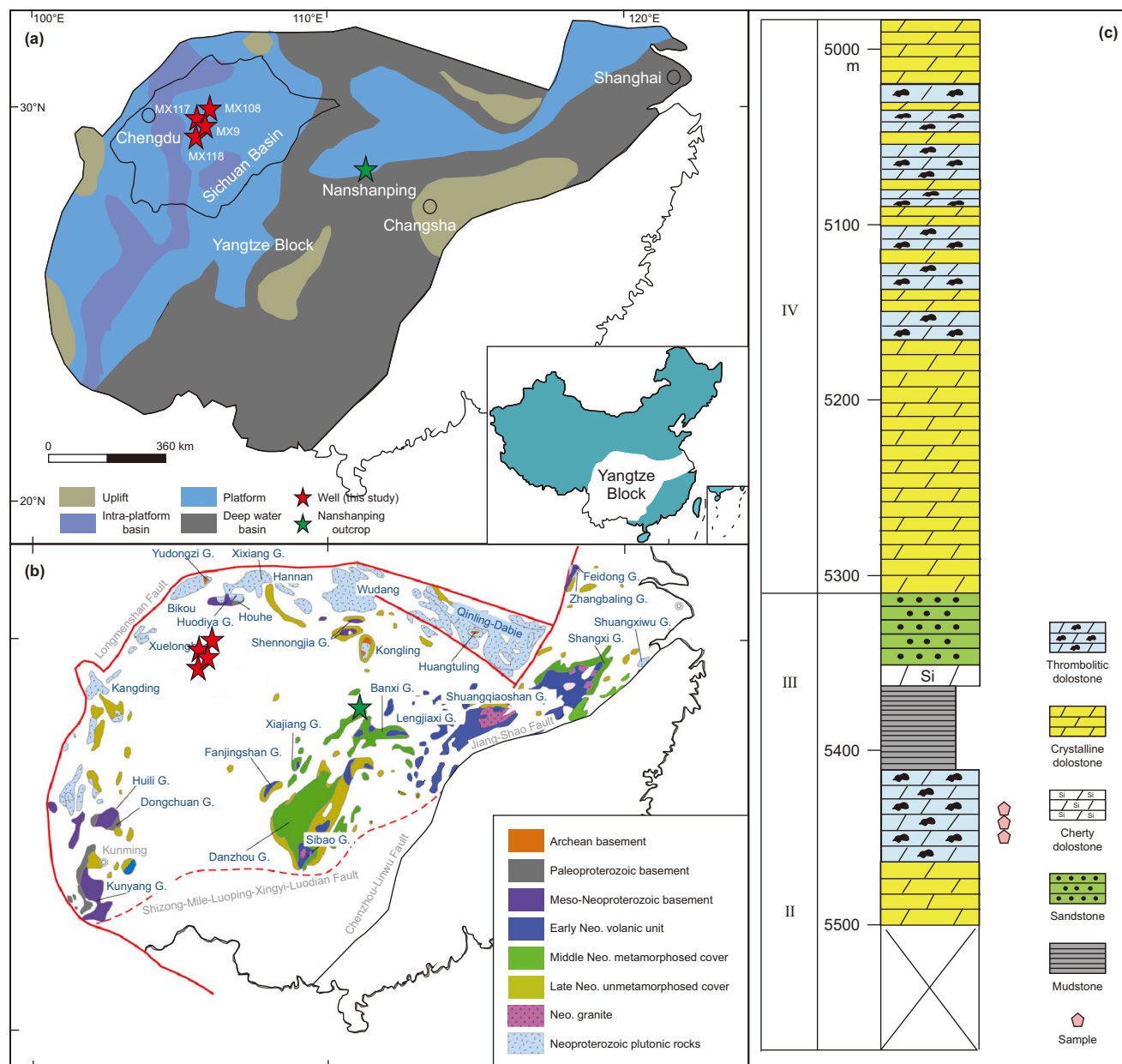


Fig. 3. Simplified map of the Yangtze Block showing the sedimentary facies and lithologies of the upper Ediacaran Dengying Formation and the Proterozoic basement. **(a)** Simplified sedimentary facies of the Yangtze Block showing the Yangtze Basin, Dolostone samples analysed here are from drilled wells and outcrops in the Sichuan Basin, upper Yangtze Block, while the Nanshanping outcrop in the middle Yangtze Block is modified after Muñoz-López et al. (2024). **(b)** Simplified map showing the Proterozoic basement in the Yangtze Block (after Zhao and Cawood (2012)). **(c)** Lithological column of the Dengying Formation from well MX9 in the Sichuan Basin. Moreover, samples from this well that were analysed for Mg and Ca isotopes are labelled in this figure.

(calcite) were routinely analysed throughout the experiments to ensure accuracy and reliability.

3.2.2. Magnesium ($\delta^{26}\text{Mg}$) isotope

For $\delta^{26}\text{Mg}$ isotopic composition analysis, ca. 0.5 mg of dolomite powder was weighed, dissolved in 6 M HCl. The solution was subsequently dried and approximately 250 μL of a mixture of HNO_3 and H_2O_2 was added. After evaporation to dryness, the samples were re-dissolved in 1.25 M HCl. Typically, 50 μg of Mg passed through the ion exchange columns. The Mg fraction was recovered using BioRad ion exchange resin AG50 W-X12 (200–400 mesh) and subsequently dried. The dried Mg fraction was re-dissolved to 3.5% HNO_3 forming a 500 ppb Mg-solution that was

measured via a Thermo Fisher Scientific Neptune multiple-collector inductively coupled plasma mass spectrometry (MC-ICP-MS). Analytical methods are described in detail in Immenhauser et al. (2010) and Riechelmann et al. (2018). The internal precision of the measurements is suggested as $\pm 2\sigma$, and the external precision was determined by measuring the mono-elemental solution Cambrian I. The magnesium isotopic values are reported relative to DSM3 standard solution. Cambridge I ($\delta^{25}\text{Mg} = -1.37\text{‰}$ (avg.) $\pm 0.02\text{‰}$ (2σ), $\delta^{26}\text{Mg} = -2.65\text{‰} \pm 0.03\text{‰}$, $n = 55$), JDo-1 dolomite standard ($\delta^{25}\text{Mg} = -1.23\text{‰} \pm 0.01\text{‰}$, $\delta^{26}\text{Mg} = -2.36\text{‰} \pm 0.03\text{‰}$, $n = 3$), and Ruhr-University Bochum limestone standard RUB limestone standard ($\delta^{25}\text{Mg} = -1.91\text{‰} \pm 0.02\text{‰}$, $\delta^{26}\text{Mg} = -3.66\text{‰} \pm 0.04\text{‰}$, $n = 6$), moreover, were

Table 1
Brief summary of parameters for the diagenetic models.

Parameter	Definition	Crustal fluid model	Mixing of seawater (80%) and crustal fluids (20%) model	Mantle-derived fluid model	Mixing of seawater (80%) and mantle-derived fluids (20%)	Reference
α_{Mg}	Mg isotopic fractionation factor	0.9993	0.9993	0.9993	0.9993	Li et al. (2015)
K _{Sr}	Distribution coefficient for Sr	0.01	0.01	0.01	0.01	Ahm et al. (2018)
M _f	Mass of element in fluid, mmol/kg	Sr = 0.09; Mg = 20	Sr = 0.09; Mg = 20	Sr = 0.05; Mg = 20	Sr = 0.07; Mg = 20	Ahm et al. (2018); Wei and Algeo (2020)
M _s	Mass of element in primary sediment, ppm	Sr = 700; Mg = 5000	Sr = 700; Mg = 5000	Sr = 700; Mg = 5000	Sr = 700; Mg = 5000	Husson et al. (2015); Ahm et al. (2019); Wei et al. (2022)
δf	Isotopic value of fluid	$^{87}\text{Sr}/^{86}\text{Sr} = 0.717$	$^{87}\text{Sr}/^{86}\text{Sr} = 0.7102$	$^{87}\text{Sr}/^{86}\text{Sr} = 0.704$	$^{87}\text{Sr}/^{86}\text{Sr} = 0.7076$	Palmer and Elderfield (1985); Salters and Stracke (2004); Huang et al. (2020)
		$\delta^{26}\text{Mg} = -1.6\text{‰}$	$\delta^{26}\text{Mg} = -0.6\text{‰}$	$\delta^{26}\text{Mg} = 0.2\text{‰}$	$\delta^{26}\text{Mg} = -0.28\text{‰}$	Teng (2017); Kimmig et al. (2021); Mavromatis et al. (2021)
δs	Isotopic value of primary sediment	$^{87}\text{Sr}/^{86}\text{Sr} = 0.7085$	$^{87}\text{Sr}/^{86}\text{Sr} = 0.7085$	$^{87}\text{Sr}/^{86}\text{Sr} = 0.7085$	$^{87}\text{Sr}/^{86}\text{Sr} = 0.7085$	Palmer and Elderfield (1985); Salters and Stracke (2004); Huang et al. (2020); El Meknassi et al. (2018)
		$\delta^{26}\text{Mg} = -3\text{‰}$	$\delta^{26}\text{Mg} = -3\text{‰}$	$\delta^{26}\text{Mg} = -3\text{‰}$	$\delta^{26}\text{Mg} = -3\text{‰}$	Li et al. (2024)

Table 2
Elemental concentrations, $\delta^{26}\text{Mg}$ and $\delta^{44/40}\text{Ca}$ values of saddled dolomite from the upper Ediacaran Dengying Formation, Sichuan Basin. na = sample not analysed.

Well	Depth, m	Mg/Ca molar ratios	Sr, ppm	Fe, ppm	Mn, ppm	$\delta^{25}\text{Mg}$, ‰ (DSM3)	$\pm 2\sigma$	$\delta^{26}\text{Mg}$, ‰ (DSM3)	$\pm 2\sigma$	$\delta^{44/40}\text{Ca}$, ‰ (SRM915a)	$\pm 2\sigma$
MX117	5217.55	0.99	53.5	69.5	140.0	-1.08	0.01	-2.12	0.01	na	na
MX108	5316.64	0.94	157.2	49.5	232.0	-1.16	0.01	-2.23	0.01	1.10	0.13
MX118	5132.48	1.00	33.8	648.6	553.0	-1.21	0.03	-2.33	0.03	na	na
MX9	5428.4	0.99	62.8	318.3	719.0	-0.98	0.02	-1.90	0.02	na	na
MX9	5433.05	0.99	135.8	274.2	543.0	-1.24	0.01	-2.40	0.02	na	na
MX9	5441.74	0.99	180.3	303.6	410.0	-1.24	0.01	-2.41	0.03	0.81	0.03

measured to test the reproducibility and yielded similar results compared to other laboratories (Immenhauser et al., 2010; Riechelmann et al., 2016, 2018).

3.2.3. Calcium ($\delta^{44/40}\text{Ca}$) isotope

For calcium ($\delta^{44/40}\text{Ca}$) isotopic analysis, 0.05–0.15 mg of dolomite powder was weighed, dissolved in 1 mL 6 M HCl, and subsequently dried at 125 °C on a hot plate. Samples were then dissolved in 1 mL 3 M HNO₃ and Sr was separated from the remaining sample using TRISKEM SrResin (100–150 mesh) and PFA columns. The residue was evaporated to dryness, followed by the addition of 1 mL of a 1:1 mixture of H₂O₂: HNO₃ (31%:65%) to eliminate organic compounds and prevent cation complexation. Samples were subsequently dried at 60 °C and re-dissolved in 1 mL 2.5 M HCl. The Ca fraction was separated via BioRad ion exchange resin AG50W-X12 (200–400 mesh) and quartz glass columns. Analytical methods are described in detail in Riechelmann et al. (2020) and Hoffmann et al. (2021). A 3 ppm Ca-solution prepared using HNO₃ was performed for calcium isotopic analysis via a Thermo Fisher Scientific Neptune MC-ICP-MS. The standard sample bracketing technique was performed to calculate $\delta^{44/42}\text{Ca}$ and IAPSO seawater is utilised as the standard solution. SRM-915a ($\delta^{44/42}\text{Ca} = -0.94\text{‰} \pm 0.04\text{‰}$, $\delta^{44/43}\text{Ca} = -0.44\text{‰} \pm 0.05\text{‰}$, $n = 68$) and USGS EN-1 ($\delta^{44/42}\text{Ca} = -0.54\text{‰} \pm 0.07\text{‰}$, $\delta^{44/43}\text{Ca} = -0.28\text{‰} \pm 0.08\text{‰}$, $n = 5$), moreover, were measured to test the reproducibility and showed similar results compared to other laboratories (Hippler et al., 2013; Riechelmann et al., 2020).

For comparison with published data, the $\delta^{44/42}\text{Ca}$ values measured here are converted into $\delta^{44/40}\text{Ca}$ values using the equation from Gussone et al. (2016) in the context of mass-dependent fractionation.

$$\delta^{44/40}\text{Ca} \approx \delta^{44/42}\text{Ca} \times \left[\ln(m^{44}\text{Ca} / m^{40}\text{Ca}) / \ln(m^{44}\text{Ca} / m^{42}\text{Ca}) \right] \quad (1)$$

where $m^x\text{Ca}$ represents the atomic mass of each Ca isotope. The conversion of the IAPSO standard to SRM 915a was performed using the following equation from Gussone et al. (2016):

$$\delta^{44/40}\text{Ca}_{\text{SRM 915a}} = \delta^{44/40}\text{Ca}_{\text{IAPSO}} + 1.88 \quad (2)$$

3.3. Numerical modelling

A numerical model was performed to constrain the evolution of carbonate's geochemical signatures in the context of hydrothermal alteration in general. This model simulates replacement dolomitisation by transforming the limestone into a dolostone lithology through dissolution and re-precipitation (e.g., Ahm et al., 2018, 2019). The model represents a one-dimensional system of 15 boxes with volume of 1 m³, where fluids move through the sediment column from the uppermost box to the next via advection. Along this unidirectional flow path, the system evolves

progressively from a fluid-buffered to a sediment-buffered condition (modified after Ahm et al. (2018)). The elemental concentrations, isotopic compositions, and fractionation factors of the primary limestone and fluids, together with model parameters and related references, are listed in Table 1 and S1. The time-dependent variations in the isotopic compositions of the bulk carbonates consist of the relative fraction of primary and secondary minerals within the sediment (Ahm et al., 2018; Wei et al., 2019):

$$\frac{\partial M_f \delta_f}{\partial t} = -u \frac{\partial M_f \delta_f}{\partial x} + R \left[M_{\text{prim}} \delta_{\text{prim}} - m C_{\text{prim}} (\delta_f + 10^3 \ln \alpha) \right] \quad (3)$$

where M_f is the mass of a specific element in the fluid within the pore-space of one box (e.g. Mg, Sr), and δ_f is the isotopic composition of the fluid. M_{prim} and δ_{prim} is the mass and isotopic composition of the primary mineral phase. C_{prim} is the mass of carbon in the primary mineral, while a and u are isotopic fractionation factor and constant advection rate, respectively. To conserve mass of carbon in the sediment during diagenesis, m is used here as a stoichiometric ratio to scale the precipitation rate ($R \times C_{\text{prim}} \times m$) of a specific element relative to carbon (e.g. for Mg in dolomite; Mg/C = 0.5).

The spatial dimension (∂x) is discretized via finite differences so that the time-dependent differential equation is reduced to an ordinary differential equation that is solved numerically for each box (n). This equation shows the evolution of the fluid composition along a single flow path (from box 1 to n), controlling the composition of the precipitating diagenetic mineral in each box (please refer to Ahm et al. (2018) for more details).

$$\frac{\partial M_f^n \delta_f^n}{\partial t} = -u \left[M_f^{n-1} \delta_f^{n-1} - M_f^n \delta_f^n \right] + R \left(M_{\text{prim}}^n \delta_{\text{prim}}^n - m C_{\text{prim}}^n \delta_{\text{sec}}^n \right) \quad (4)$$

4. Results

4.1. Petrology

Lithofacies (mainly host dolostone) in the Dengying Formation consist predominantly of zebra (fabric) dolostone, crystalline dolostone, and microbial dolostone. Zebra dolostone is characterised by bedding-parallel, elongated (several dm), 1–2 cm wide pores (dissolution voids) pinching out laterally. These are embedded in grey matrix dolostone, resulting in an “ABBA” sequence (A: hostrock; B: saddle dolomite; Fig. 4(a) and (b); Vandeginste et al., 2005; Peng et al., 2018). The open, elongated pores that are typical of zebra fabric dolostone are partially filled by white saddle dolomite, pyrite, quartz and fluorite. The crystalline dolostone is dark grey to grey in hand specimens and is composed of dolomite crystals with sizes ranging from 10 to 100 μm (Hu et al., 2023a). Microbial dolostone predominantly consists of dolomitic stromatolites and dolomitic thrombolites (Fig. 4(c)). Dolomitic stromatolites display distinct translucent and dark-pigmented laminae, while thrombotic texture consists of micrometre to centimetre-scale irregular to rounded microclots.

Saddle dolomite (Machel and Lonnee, 2002; Zhang et al., 2024) is one of the most predominant pore-filling phases in the Dengying Formation (Fig. 4(d)). Saddle dolomite overgrows medium-to coarse-crystalline dolomite cement and is the last dolomite phase in vugs or fractures. This phase is composed of nonplanar (warped) to subhedral crystals with dimensions ranging from 20 to 2500 μm . Saddle dolomites show undulatory extinction under cross-polarised light. Under cathodoluminescence, bright red to dull red zones with patchy appearance (Fig. 4(e)) or mottled dull-to bright-red color are presented. Saddle dolomite shows a

spatially close relationship to pyrite, fluorite (Fig. 4(d)), quartz and calcite cement (Hu et al., 2020b). Other types of dolomite cement include fibrous dolomite cement, fine-crystalline dolomite cement, and medium-to coarse-crystalline dolomite cement (Fig. 4(f)).

4.2. Geochemistry

Saddle dolomite in the Sichuan Basin yields Mg/Ca molar ratios of 98.3% (avg.) $\pm$ 1.9% (σ) ($n = 6$, Fig. 5, Table 2). Strontium concentrations range from 33.8 to 180.3 ppm, with an average of 103.9 ppm for saddle dolomite. This dolomite phase shows $\delta^{26}\text{Mg}$ isotopic compositions of $-2.23\text{‰} \pm 0.18\text{‰}$ ($n = 6$), and two samples yield $\delta^{44/40}\text{Ca}$ values of 0.81‰ and 1.10‰.

5. Interpretation and discussion

This work aims to evaluate different Mg sources for (replacement) dolomitisation at burial depths. This is performed by using a multi-proxy isotope record from the Dengying Formation sampled in two locations in the Yangtze Block combined with data extracted from published work. The isotope geochemistry of the upper Ediacaran hydrothermal dolomite in the Sichuan Basin, upper Yangtze Block is contrasted with published values of the upper Ediacaran hydrothermal dolomite in the Nanshanping outcrop, middle Yangtze Block. Note that the term “hydrothermal” is used in the sense that an advected fluid is at least 5 °C warmer (considerably more in the case studied dealt with here) than the reacting host rock (see White, 1957). Moreover, we apply numerical modelling to constrain the evolution of the geochemical signatures of these carbonates in the context of hydrothermal dolomitisation.

The authors recognise that each geological basin and the associated fluid within it may be unique. Consequently, it can be challenging to compare different basins, the dolostones they contain, and the fluids that contributed to their formation over time. This difficulty arises from variations in fluid properties at depth, which can include differences in fluid sources (such as meteoric, marine, connate, brine, Na-Ca-Cl-type, and crust-derived), as well as their geochemistry, temperature, and pH. Additionally, there is the consideration of whether one or multiple types of fluids mix (see Immenhauser, 2022, and the references therein for an overview). Despite these challenges, important review papers and case studies (Gomez-Rivas et al., 2014; Peng et al., 2018; Kimmig et al., 2021; Muñoz-López et al., 2024) have highlighted significant similarities that should be taken into account when comparing burial dolomites and their corresponding fluids over geological time.

Based on published work and new data, this study examines geochemical differences between palaeo-seawater and host dolostone compared to coeval hydrothermal (burial) dolomite throughout Earth's history (Fig. 2). Variance is observed and expected, as fluids at depth can significantly differ in their geochemistry from marine waters. Our own data are investigated to contextualise the sources of magnesium that led to different types of hydrothermal dolomitisation in the upper Ediacaran Sichuan Basin and the middle Yangtze Block. In the next step, these findings are placed within a broader scientific framework to develop general models for hydrothermal dolomitisation throughout geological time and beyond a given sedimentary basin.

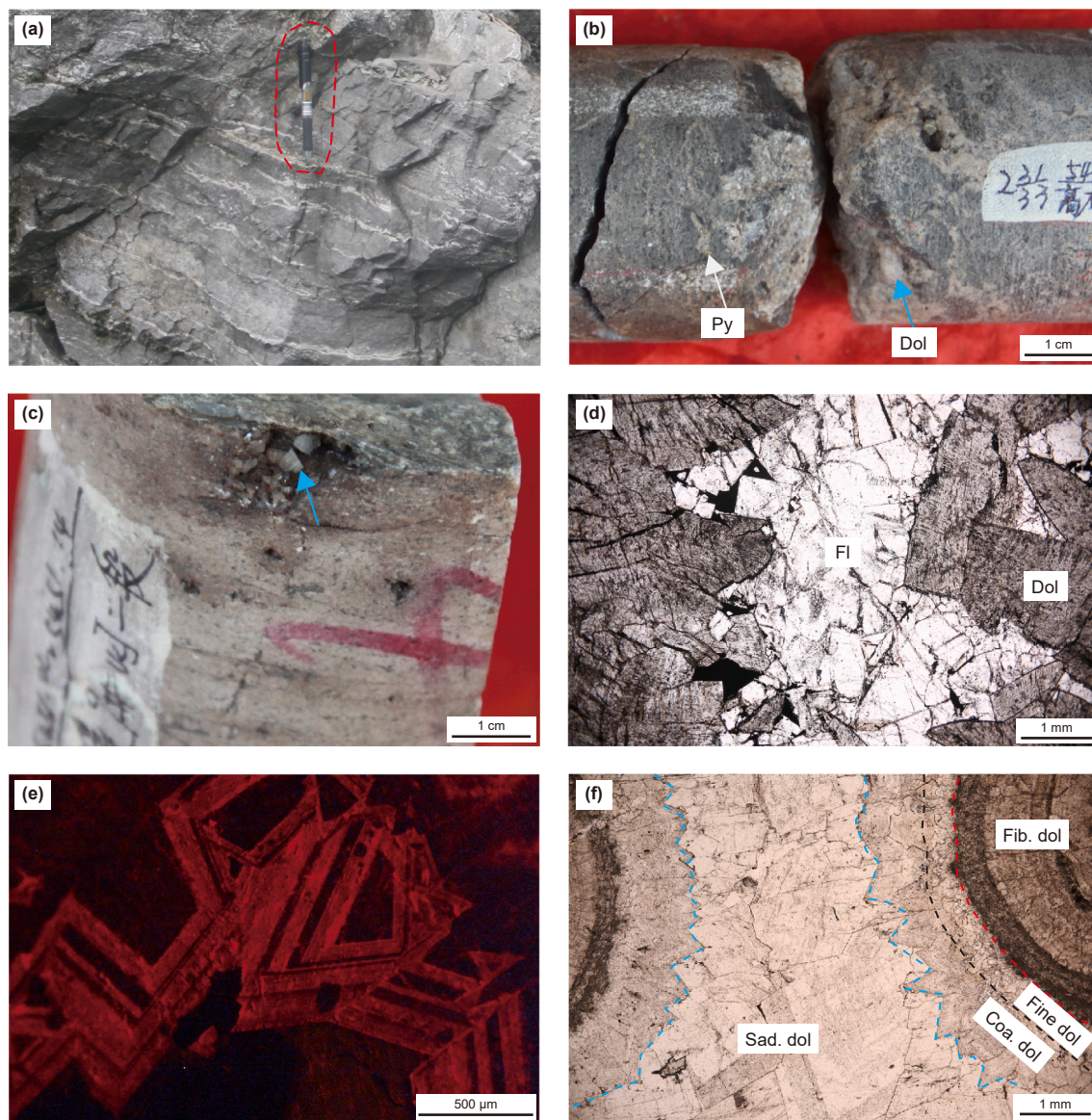


Fig. 4. Core images and thin-section photomicrographs showing zebra dolostone fabric, thrombolite and saddle dolomite in the Dengying Formation. (a) Zebra dolostone fabric, Ebian outcrop. (b) Zebra dolostone fabric with saddle dolomite (blue arrow) and pyrite (white arrow), well GS32, at a depth of 5426.75 m. (c) Thrombolite with saddle dolomite crystals (blue arrow), well MX9 at a depth of 5451.14 m. (d) Saddle dolomite (Dol) and fluorite (Fl) occur as the pore-filling fabric, well MX52 at a depth of 5572.28 m, plane-polarised light (PPL). (e) Cathodoluminescence photomicrograph showing growth zones of saddle dolomite, well GK1 at a depth of 5406 m. (f) Overgrowth of saddle dolomite on medium-to coarse-crystalline dolomite cement (Coa. Dol), fine-crystalline dolomite cement (Fine dol), and fibrous dolomite cement (Fib. Dol), well MX9 at a depth of 5457.76 m, PPL.

5.1. Constraints on the petrography of zebra dolostone fabrics and their fluid temperature in the Dengying Formation

Zebra dolomite fabrics and the typically related saddle dolomites are common burial (hydrothermal) minerals precipitated over a temperature range of some 10's to >200 °C (e.g., Davies and Smith, 2006; Hollis et al., 2017; see review and critique in Zhang et al., 2024). The upper Ediacaran saddle dolomite in the Sichuan basin overgrows medium-to coarse-crystalline dolomite cement, and is associated with typical hydrothermal phases such as fluorite and quartz (Fig. 4(d) and (f)). In the samples we studied, saddle dolomite showing elevated fluid temperatures in both the Sichuan Basin ranging from 170 to 260 °C (Liu et al., 2016a; Hu et al., 2020b) and the Nanshanping outcrop ranging from 210 to 240 °C (Muñoz-

López et al., 2024). Most of these temperatures are 5 to 10 °C higher than the maximum temperatures of approximately 210 to 220 °C that the strata experienced (Liu et al., 2016b), thus the saddle dolomite is hydrothermal (Machel and Lonnee, 2002). The saddle dolomite in Sichuan Basin yields relatively low $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values, high $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, Fe and Mn concentrations with typical middle REE enrichment compared to host dolostone and early diagenetic dolomite (Hu et al., 2020b, 2023b; Jiang et al., 2023).

Petrographic observations reveal that the zebra texture, characterised by alternating light and dark bands in the Nanshanping outcrop (Muñoz-López et al., 2024; McCormick et al., 2025), together with the patchy cathodoluminescent zonation (Fig. 4(e)) and the mottled luminescent color exhibited by certain saddle

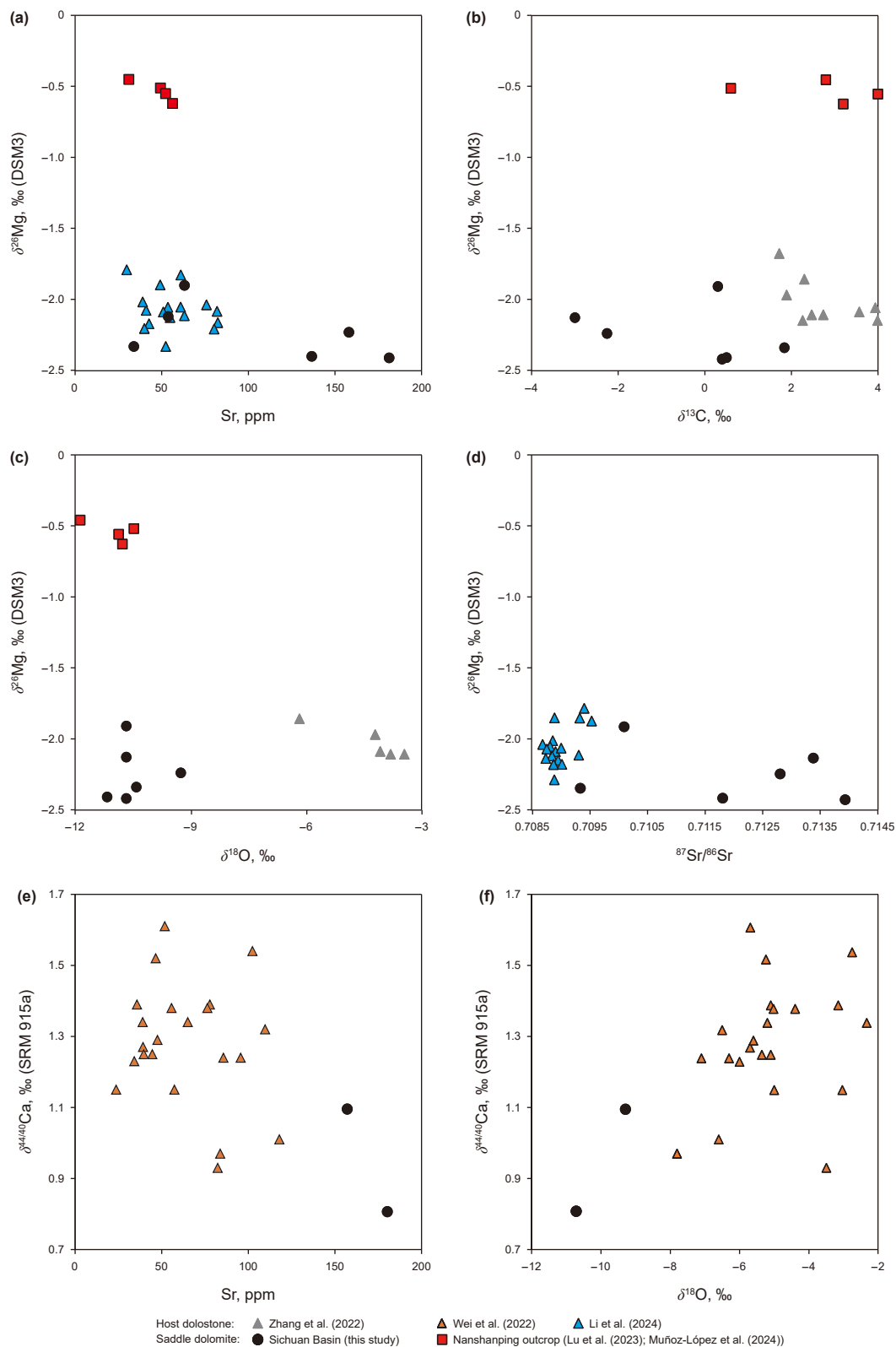


Fig. 5. Cross-plot of geochemical properties of hydrothermal/burial dolomite compared to coeval host dolostone in the Dengying Formation, Yangtze Block. **(a)** Cross-plot of $\delta^{26}\text{Mg}$ (DSM3) versus Sr contents. **(b)** Cross-plot of $\delta^{26}\text{Mg}$ (DSM3) versus $\delta^{13}\text{C}$ values. **(c)** Cross-plot of $\delta^{26}\text{Mg}$ (DSM3) versus $\delta^{18}\text{O}$ values. **(d)** Cross-plot of $\delta^{26}\text{Mg}$ (DSM3) versus $^{87}\text{Sr}/^{86}\text{Sr}$ values. **(e)** Cross-plot of $\delta^{44/40}\text{Ca}$ values (SRM 915a) versus Sr contents. **(f)** Cross-plot of $\delta^{26}\text{Mg}$ (DSM3) versus $\delta^{18}\text{O}$ values. Apart from the data analysed here, other data are collected from Wei et al. (2022), Zhang et al. (2022), Lu et al. (2024), Li et al. (2024) and Muñoz-López et al. (2024).

dolomites (Liu et al., 2021b), collectively indicate that these saddle dolomites formed through a replacement process. In addition, the variability in carbon and oxygen isotopic compositions between matrix and saddle dolomites further supports the replacement of marine host carbonates by hydrothermal dolomite, as previously documented in the Sichuan Basin (Feng et al., 2017).

5.2. The Dengying formation case study: implications for Mg source fingerprinting

5.2.1. $^{87}\text{Sr}/^{86}\text{Sr}$ ratio and elemental geochemistry

In the cross-plot of different geochemical parameters (i.e., Sr contents, $\delta^{13}\text{C}$ values, $\delta^{18}\text{O}$ values, and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, Fig. 5), the upper Ediacaran hydrothermal dolomite data are set apart from host dolostone and show complex patterns. Furthermore, saddle dolomite proxy data from the upper Yangtze Block differ markedly from those in the middle Yangtze Block. These lines imply that several types of hydrothermal fluid were involved. In the following, the different Mg sources are evaluated.

The absence of Ediacaran or older sulphates in the Yangtze Block eliminates the possibility that dissolved evaporites are the main source of magnesium. Additionally, the intricate geochemical patterns of basin-scale hydrothermal dolomitisation observed here contrast with the previously suggested model, which proposed that crustal fluids are the single source of Dengying dolomites in the Yangtze Block (Jiang et al., 2016; Feng et al., 2017; Muñoz-López et al., 2024). Both crustal and mantle-derived fluids likely accounted for hydrothermal dolomitisation here.

Compared to the upper Ediacaran host dolostone (Sr: 40 to 80 ppm, $^{87}\text{Sr}/^{86}\text{Sr}$ ratios: 0.7087 to 0.7094, Hu et al., 2020b; Li et al., 2024), saddle dolomite from the Sichuan Basin yields significantly high Sr contents of 103.9 ± 56.0 ppm (Fig. 5(a)) and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of 0.7119 ± 0.0017 (Fig. 5(d)–Hu et al., 2023a). In contrast, saddle dolomite from the Nanshanping outcrop has low Sr contents of 47.0 ± 9.6 ppm (Lu et al., 2024). These data are consistent with the geochemical characteristics of crust and mantle rocks, respectively. Previous studies (Palmer and Elderfield, 1985; Salters and Stracke, 2004) show that mantle rocks are characterised by low Sr concentrations (e.g., 9.8 ppm for depleted mantle) and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (0.7035). The Nanshanping outcrop (Fig. 3(b)) lies near igneous rocks, products of mantle plume activity during the Rodinia break-up (Li et al., 2003; Zhao and Cawood, 2012; Yao et al., 2019). Hydrothermal fluids migrated along faults, interacted with these mantle rocks, and later penetrated the Dengying Formation. In contrast, continental crust (sedimentary rocks are included), such as shale and chlorite minerals, yield high Sr concentrations and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of 85–400 ppm and 0.7400, respectively (Huang et al., 2020; Wei and Algeo, 2020; Lenz et al., 2024). The relatively low Sr content (<100 ppm) and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (0.7070 to 0.7200) of magnesite deposits, potentially present at depth, suggest that this mineral is not a likely magnesium source (Frimmel and Niedermayr, 1991; Kuznetsov et al., 2007) in the Sichuan Basin. Correspondingly, the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of saddle dolomite in the Sichuan Basin show a decreasing trend along the pathway of hydrothermal fluids, resulting from the decreasing proportion of continental crustal fluids in the mixing process (Hu et al., 2023a).

Notably, saddle dolomite in the Sichuan Basin yields relatively lower $\delta^{13}\text{C}$ values ($-0.4\text{‰} \pm 1.7\text{‰}$) compared to those ($2.5\text{‰} \pm 1.5\text{‰}$, Lu et al., 2024) from the Nanshanping outcrop and their host dolostone in Yangtze Block (Fig. 5(a)). This is ascribed to CO_2 generated via thermochemical sulphate reduction in the Dengying Formation in the Sichuan Basin (Hu et al., 2020b). Alternatively, the incorporation of organic matter

in crustal fluids may result in low $\delta^{13}\text{C}$ signatures (Walker et al., 2008). Both would further support the involvement of crust-derived fluids in the dolomitisation process in the Sichuan Basin. From the perspective of $\delta^{18}\text{O}$ values, saddle dolomite in the Sichuan Basin ($-10.5\text{‰} \pm 0.6\text{‰}$, Fig. 5(c)) yields comparable values to those from Nanshanping outcrop ($-10.9\text{‰} \pm 0.5\text{‰}$, Lu et al., 2024). This might be related to their similar fluid temperature as discussed in section 5.1.

In conclusion, based on $^{87}\text{Sr}/^{86}\text{Sr}$ ratio and elemental geochemistry, the following pattern emerges: for the case example of the Sichuan Basin, the contribution of continental crustal fluids leads to relatively high Sr concentrations and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of burial dolomite. In the case of the Nanshanping outcrop, burial dolomite geochemistry is in agreement with the involvement of mantle-derived fluids.

5.2.2. Magnesium ($\delta^{26}\text{Mg}$) isotopic archives

Magnesium ($\delta^{26}\text{Mg}$) isotopic values bear the potential to identify dolomitisation fluids and trace dolomitisation history (Walter et al., 2015; Muñoz-López et al., 2024). That said, the complexity of burial dolomitisation and Mg cycling is remarkable (Geske et al., 2015a, 2015b; Li et al., 2023, 2024), and hence, significant omissions still exist. Compared to the Upper Ediacaran host dolostone in both settings (-2.2 to -1.8‰ , Li et al., 2024), saddle dolomite in the Sichuan Basin yield ^{26}Mg -depleted values of $-2.23 \pm 0.18\text{‰}$. In contrast, saddle dolomite collected from the Nanshanping outcrop (Fig. 2(a)) shows ^{26}Mg -enriched values ranging from -0.6‰ to -0.5‰ (Fig. 5(a)–Table S2, Muñoz-López et al., 2024). This feature merits discussion.

We follow previous workers and suggest that differences in dolomite $\delta^{26}\text{Mg}$ isotopic composition might be caused by the following parameters: fluid source and temperature, formation/precipitation rate (disequilibrium), aqueous Mg isotopic composition ($\delta^{26}\text{Mg}_{\text{aq}}$) and variable fractionation factor between dolomite and aqueous ($\Delta^{26}\text{Mg}_{\text{dolo-aq}}$; Immenhauser et al., 2010; Huang et al., 2015; Teng, 2017; Riechelmann et al., 2020, 2022). Other factors, such as mineralogy (Teng, 2017), microbial activity (Riechelmann et al., 2020), meteoric alteration (Akhtar et al., 2024), and near-surface weathering (Kasemann et al., 2014), are excluded due to the intermediate-to-deep burial diagenetic environment in which hydrothermal dolomitisation occurs shown here (Jiang et al., 2023).

Temperature-dependent function for Mg isotope fractionation between hydrothermal dolomite and aqueous is $\Delta^{26}\text{Mg}_{\text{dolo-aq}} = -0.1554 (\pm 0.0096) \times 10^6/T^2$ (Li et al., 2015). However, saddle dolomite samples from the Sichuan Basin and the Nanshanping outcrop are formed at a high-temperature range of 170–260 °C (Liu et al., 2016a; Hu et al., 2020b) and 210–240 °C (Muñoz-López et al., 2024), respectively. This is relevant, as temperature-induced fractionation between carbonate and its aquatic environment is subdued or nil in high-temperature burial environments (Geske et al., 2012; Lavoie et al., 2014). Furthermore, the temperature range shown here yields $\delta^{26}\text{Mg}$ differences of 0.25‰ using fractionation equation of Li et al. (2015), lower than the significant range of saddle dolomite in the Dengying Formation. In contrast, previous studies (Immenhauser et al., 2010; Huang et al., 2015) document that at higher precipitation rates, Mg isotopic fractionation between the carbonate phase and its precipitating fluid is observed. That said, these effects might be minimal due to their similar burial fluid temperatures of about 200 °C (and higher).

As a working hypothesis, it is proposed that different magnesium sources are likely responsible for the variant $\delta^{26}\text{Mg}$ values of upper Ediacaran saddle dolomite (Fig. 5). Variable degrees of mixing between burial brines (or heated seawater) and exotic fluids exert significant control on the isotopic characteristics of

parent fluids (Geske et al., 2015a; Wei et al., 2019). Applying fractionation functions between hydrothermal dolomite and its ambient fluid (Li et al., 2015) and simplified formation temperature of 200 °C (Liu et al., 2016a; Hu et al., 2020b; Muñoz-López et al., 2024), dolomitising fluids result that have averaged magnesium isotope compositions of -1.7‰ to -1.2‰ (Sichuan Basin) and -0.01‰ to 0.2‰ (Nanshanping outcrop), respectively. The former values fall into the range between coeval host dolostone (Fig. 2(a)) and the continental crust, which is characterised by a large range of $\delta^{26}\text{Mg}$ values with low values for some Mg-bearing minerals, such as magnesite (-2.95‰ , Mavromatis et al., 2021), chlorite (-1.8‰ , Kimmig et al., 2021), crustal granite and shale (-1.64‰ , Teng, 2017), respectively. Moreover, chlorite tends to release predominantly ^{24}Mg during the initial dissolution stage (Ma et al., 2015). This is relevant as Ma et al. (2019) documented that Permian extensional tectonics induced large volumes of chlorite (12%–30%) in igneous rocks of the Sichuan Basin. In addition, microbial dolostone is widely developed in the Dengying Formation of the Sichuan Basin (Zhu et al., 2022). Microbially induced dolostone is likely characterised by relatively low $\delta^{26}\text{Mg}$ values (Geske et al., 2015b; Zhang et al., 2022; Qiao et al., 2023), which may have been transferred to the aqueous fluids through interactions between hydrothermal fluids and microbial dolostone. Such fluid–rock interactions likely resulted in a decrease in the $\delta^{26}\text{Mg}$ values of the hydrothermal fluids, thereby contributing to the formation of hydrothermal dolomite with low $\delta^{26}\text{Mg}$ values in the Sichuan Basin.

The Nanshanping outcrop is spatially close to the Jiangnan Orogenic Belt (Fig. 3(b)). There, a mantle superplume formed during the breakup of the supercontinent Rodinia (Li et al., 2003; Yao et al., 2019). Consequently, the magnesium isotope compositions of dolomitising fluids estimated of the Nanshanping outcrop saddle dolomites are within the average mantle $\delta^{26}\text{Mg}$ values (-0.25‰ , Teng, 2017). Crust- and mantle-derived fluids are magnesium-rich and are highly reactive, releasing magnesium

with typical values into hydrothermal fluids (Hansen et al., 2005; Muñoz-López et al., 2024). It seems likely that ascending mantle/crust-derived fluids are mixed with burial brine (or modified seawater) at depth. Correspondingly, hydrothermal dolomite yields geochemical archives falling into the range of host dolostone to mantle/crust. In the authors' view, this range in fluid types and various degrees of mixing has caused the complex isotope signatures observed (Fig. 6).

In summary, hydrothermal fluids interacted from mantle and crustal levels (e.g., chlorite-derived Mg) and then ascended upward (buoyancy-driven), mixing with brine, into the Dengying Formation of the upper Yangtze Block. Correspondingly, the saddle dolomites at this site are characterised by ^{26}Mg -depleted values but relatively high Sr contents and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios. In contrast, the ^{26}Mg -enriched values and low Sr contents of the coeval saddle dolomite in the middle Yangtze Block might be related to the involvement of mantle-derived fluids.

5.2.3. Calcium ($\delta^{44}/^{40}\text{Ca}$) isotopic archives

The upper Ediacaran saddle dolomite exhibits depleted $\delta^{44}/^{40}\text{Ca}$ values of 0.81‰ and 1.1‰ (Fig. 5(e) and (f)), which contrasts with the coeval seawater value of 1.28‰ to 1.50‰ (Ahm et al., 2019; Wei et al., 2019) and the value of around 1.2‰ for host dolostone (Wei et al., 2022). In comparison to host dolostone and early diagenetic dolomite, reports of late diagenetic or hydrothermal dolomite $\delta^{44}/^{40}\text{Ca}$ values are relatively scarce in the literature (Steuber and Buhl, 2006; Perez-Fernandez et al., 2017).

The difference in $\delta^{44}/^{40}\text{Ca}$ signatures between host dolostone and burial saddle dolomite might be temperature-induced. Aqueous Ca isotopic compositions ($\delta^{44}/^{40}\text{Ca}_{\text{aq}}$) and fractionation factors ($\Delta^{44}/^{40}\text{Ca}_{\text{dolo-aq}}$) based on previous studies (Blättler et al., 2015; Bradbury and Turchyn, 2018; Gussone et al., 2016; Gong et al., 2023) provide evidence for this rationale. Due to kinetic effects, a decreasing Ca fractionation factor is correlated with increasing temperature, similar to what is reported for $\Delta^{26}\text{Mg}_{\text{dolo-aq}}$

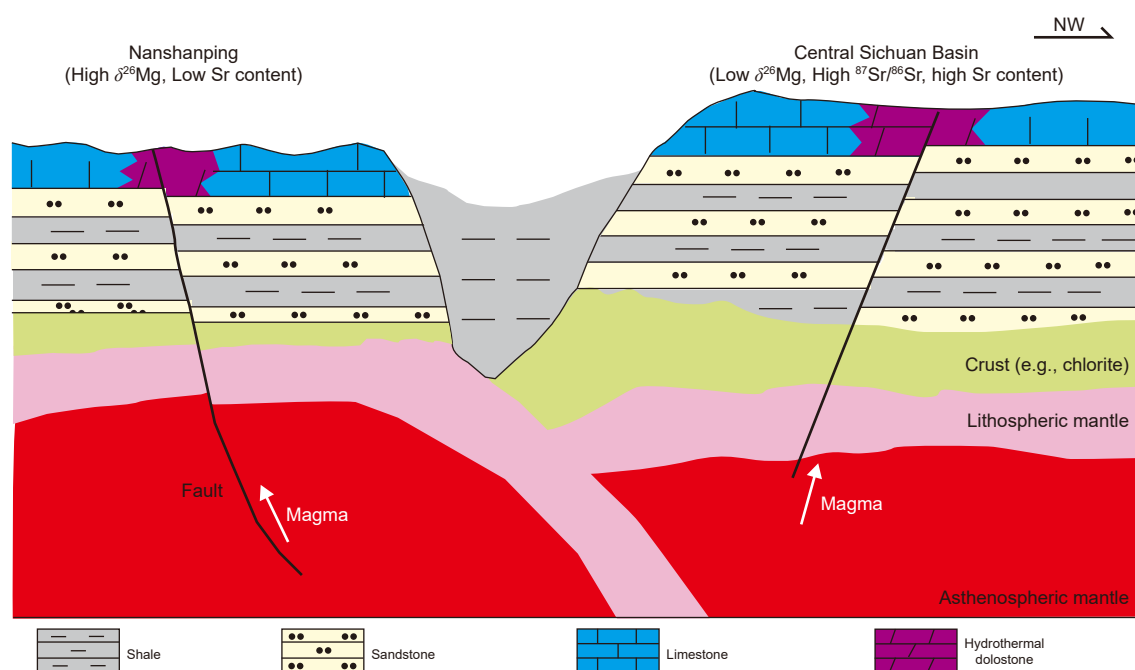


Fig. 6. Cartoon showing the precipitation of hydrothermal dolomite in the upper Ediacaran Dengying Formation. Note differences in burial dolomite $\delta^{26}\text{Mg}$ values in the Sichuan Basin (^{26}Mg -enriched, low Sr concentrations) and the Nanshanping outcrop (^{26}Mg -depleted, high Sr concentrations, $^{87}\text{Sr}/^{86}\text{Sr}$ ratios). White arrows indicate H_2O generated from the magma chamber interacting with basement rocks and migrating upwards into the Dengying Formation along fault zones, mixing with burial brine. The underlying basement is modified by Li et al. (2003), He et al. (2017) and Yao et al. (2019).

values (Gussone et al., 2016). That said, this notion is not in agreement with the lower $\delta^{44/40}\text{Ca}$ values for high-temperature saddle dolomite compared to host dolostone.

More likely, host dolostone and saddle dolomite formed in specific burial environments and stabilised in the presence of fluids with different geochemical properties and temperatures. The bulk dolostones are likely replacement dolomites that, in part, formed as non-stoichiometric phases near the seafloor, but then, replacement dolomitisation continued into the marine burial domain (see Immenhauser, 2022 for discussion). A wide range of values from $\Delta^{44/40}\text{Ca}_{\text{dolo-aq}} = -0.4\text{‰}$ to 0‰ are suggested in previous studies (Fantle and Tipper, 2014; Blättler et al., 2015; Perez-Fernandez et al., 2017). For the sake of simplicity, a $\Delta^{44/40}\text{Ca}_{\text{dolo-aq}} = 0\text{‰}$ is used here (c.f., Riechelmann et al., 2020) for burial dolomite formation (160–260 °C). Dolomite data suggest that the parent fluids likely had $\delta^{44/40}\text{Ca}$ values ranging from 0.81‰ to 1.1‰. These values fall into a range between chlorite (0.78‰ to 0.96‰, Hindshaw et al., 2014) and late Ediacaran seawater (1.28‰ to 1.50‰, Ahm et al., 2019; Wei et al., 2019).

Moreover, previous studies (Ockert et al., 2013) also show that light Ca^{2+} in a seawater or porewater environment preferentially adsorbs to clay minerals. This is further supported by case studies (Steuber and Buhl, 2006; Riechelmann et al., 2020; Chen et al., 2023) that documented lower $\delta^{44/40}\text{Ca}$ values of fluids interacting with crustal clastic bedrock, while higher values are observed for fluids percolating through carbonates.

In previous studies (Li et al., 2019; Lehn and Jacobson, 2015), the weathering of K-rich crustal rocks is suggested to supply excess ^{40}K produced by the radioactive decay of ^{40}K , thereby lowering carbonate $\delta^{44/40}\text{Ca}$ values. However, $\delta^{44/42}\text{Ca}$ values were measured in this study and then converted into $\delta^{44/40}\text{Ca}$ values. This line likely excludes the radioactive decay of ^{40}K in the hydrothermal dolomitisation in the Dengying Formation.

In conclusion, the authors believe that the interaction between hydrothermal fluids and chlorite-bearing crustal lithologies induced depleted $\delta^{44/40}\text{Ca}$ values, resulting in the precipitation of saddle dolomite with low $\delta^{44/40}\text{Ca}$ values in the Sichuan Basin (Fig. 6). This viewpoint is consistent with the understanding obtained from other geochemical proxies, including elemental contents, $\delta^{26}\text{Mg}$ values and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios. However, this study acknowledges that this finding requires further validation owing to the limited $\delta^{44/40}\text{Ca}$ dataset presented herein. Moreover, $\delta^{44/40}\text{Ca}$ values of dolomite sourced from mantle fluids merit further investigation by performing geochemical analyses in other basins. Compared with previous studies on the Dengying Formation in the Sichuan Basin (Jiang et al., 2016; Feng et al., 2017; Hu et al., 2023a) and Nanshanping outcrop (Muñoz-López et al., 2024), this study provides the first comprehensive comparison of Mg and Ca isotopic behaviours between hydrothermal and marine dolomites. Moreover, the potential sources of Mg^{2+} for hydrothermal dolomitisation are further elucidated, indicating crust-derived fluids in the Sichuan Basin and mantle-derived fluids at the Nanshanping outcrop, respectively.

5.3. General model of hydrothermal dolomitisation

A significant proportion of the dolomite found in Earth's rock record originates from secondary processes (Petrash et al., 2017; Mueller et al., 2020; Li et al., 2025). This includes recrystallised dolomicrite, which is a non-stoichiometric, early marine-diagenetic dolomite phase that often undergoes further ordering during burial. In many cases, replacement dolomite forms through the dolomitisation of sedimentary diagenetic precursor

carbonates, such as aragonite, high-Mg calcite, and low-Mg calcite. However, primary precipitates, such as saddle dolomite cement that occludes pore space, may also be common. To understand the complex patterns recorded in these carbonates, it is essential to identify the sources of magnesium in the dolomitising fluid, as this information is crucial for understanding the mechanisms of dolomite formation. Nevertheless, the types and properties of magnesium sources for late diagenetic dolomite found in deep-burial environments are intricate (Jiao et al., 2023; Muñoz-López et al., 2024).

A diagenetic model (modified from Ahm et al. (2018)) is applied to shed light on the hydrothermal dolomitisation of ancient carbonate rocks. This model simulates the replacement dolomitisation of a limestone lithology through dissolution and re-precipitation by sediment-fluid exchange reactions. In the presence of major fault zones that act as flow conduits, hydrothermal fault-zone dolomitisation is considered to create a fluid-buffered (or open) system for magnesium and strontium (Geske et al., 2015a; Muñoz-López et al., 2024). Based on the Dengying Formation and other fault-controlled hydrothermal studies (e.g., Araoka et al., 2022; Li et al., 2023), several types of hydrothermal fluids are stimulated to characterize the potential geochemical evolution of hydrothermal dolomite. Fluid types include (i) crustal fluids, (ii) mixing between crustal fluids and heated seawater with percentage of 20% and 80%, respectively, (iii) mixing between heated seawater and mantle-derived fluids with percentage of 80% and 20%, respectively, and (iv) mantle-derived fluids. Fluid mixing follows a mass-balance relationship that quantifies the proportional contributions of different sources. Linear mixing models are commonly applied to partition two end-member fluids with distinct isotopic compositions, reflecting source mixing rather than isotopic fractionation (Hoefs, 2009).

This study acknowledges that the geochemistry of crust- and mantle-derived fluids exhibits pronounced spatial and temporal variability from the Ediacaran to Phanerozoic. For simplicity and model applicability, representative end-member values were adopted here to capture the first-order geochemical distinctions between these fluid sources. Based on the Ediacaran to Phanerozoic cases (Pearce et al., 2015; Teng, 2017; Tang et al., 2020) about hydrothermal fluids at mid-ocean ridge, $\delta^{26}\text{Mg}$ values and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios are assumed to be 0.2‰ and 0.7040 of mantle-derived fluids, respectively. Referring to previous studies dealing with crust and related hydrothermal fluids (Palmer and Elderfield, 1985; Salters and Stracke, 2004; Huang et al., 2020; Mavromatis et al., 2021), $\delta^{26}\text{Mg}$ values and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of crust-derived fluids are suggested to be -0.6‰ and 0.7170, respectively. For example, ascending crustal fluids were estimated to have $\delta^{26}\text{Mg}$ value of -0.65‰ for the alteration of dolomite in the Williston Basin (Kimmig et al., 2021). Moreover, the compiled $\delta^{26}\text{Mg}$ values of hydrothermal dolomite have predominantly focused on the Ediacaran to Permian (Fig. 2(a)). For simplicity, the palaeo-seawater $\delta^{26}\text{Mg}$ value is conventionally assumed to be -0.4‰ during this period (Xia et al., 2024). Accordingly, the $\delta^{26}\text{Mg}$ value of the sediment precursor is inferred to be -3‰ , assuming a fractionation factor of 2.6‰ relative to the coexisting aqueous fluid, although Mg isotopic fractionation in carbonates spans a wide range (Immenhauser et al., 2010; Huang et al., 2015). During the Ediacaran to Permian, the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of seawater carbonates predominantly range from 0.7080 to 0.7090. Accordingly, a representative value of 0.7085 is assigned to the sediments for modelling simplicity (El Meknassi et al., 2018). The Mg contents of sediments in the model (Table 1) are consistent with those reported in the previous numerical model for Ediacaran carbonates

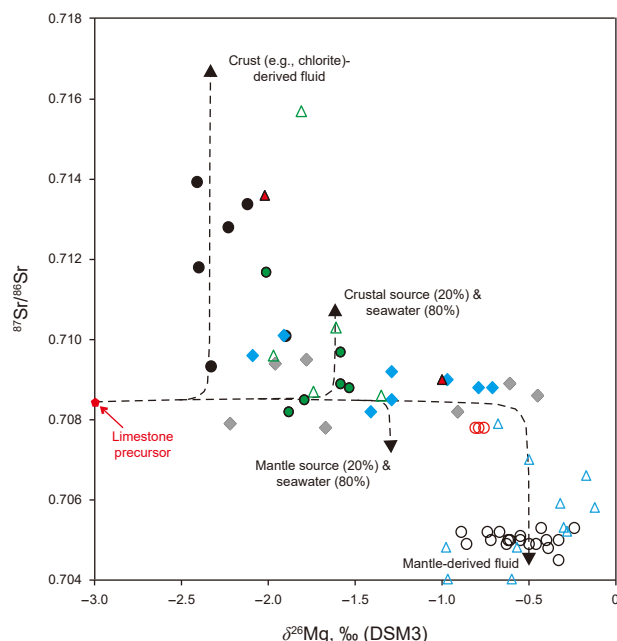


Fig. 7. Model for coupled variations of $\delta^{26}\text{Mg}$ values (DSM3) and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios for burial dolomites over the past 600 Myr. Dashed lines indicate modelled changes in $\delta^{26}\text{Mg}$ values and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios during fluid-buffered fault-zone hydrothermal dolomitisation under different conditions. The dolomitising fluids shown here include chlorite-bearing crustal sources, mixing between crustal fluids (20%) and seawater (80%), mantle-derived sources, and the mixing between mantle-derived fluids (20%) and seawater (80%). Refer to Fig. 3 for the key to symbols and Table 1 and S1 for modelling parameters.

by Ahm et al. (2019). Sr contents of sediments in the model are modified following the measured Ediacaran limestone samples (Husson et al., 2015) and numerical model of Wei et al. (2022).

Consistent with the above discussion, hydrothermal fault-zone dolomitisation likely develops a fluid-buffered (open) system for Mg and Sr, as major faults acted as conduits for fluid flow (Geske et al., 2015a; Muñoz-López et al., 2024). Accordingly, fluid-buffered trajectories of $\delta^{26}\text{Mg}$ values and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios under different hydrothermal fluid conditions are illustrated in Fig. 7. Moreover, Fig. S1 shows both fluid- and sediment-buffered variations of $\delta^{26}\text{Mg}$ values and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios. Based on our model, $\delta^{26}\text{Mg}$ values and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of hydrothermal dolomite phase show co-variations in conjunction with different types of hydrothermal fluids and the corresponding magnesium sources. In the field of crustal fluid-dominated settings, the hydrothermal dolomite phase evolves from a host dolostone towards increasing $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and decreasing $\delta^{26}\text{Mg}$ values. This scenario forms a good working hypothesis for the hydrothermal dolomite in the upper Ediacaran in the Sichuan Basin, the Cambrian Qulitage Formation in the Tarim Basin (Li et al., 2023), the Cambrian in Mississippi (Geske et al., 2015a), the Lower Ordovician in eastern Laurentia (Azmy et al., 2013), the Lower Silurian in Sayabec-West (Lavoie et al., 2014), and the Devonian Winnipegosis Formation in Williston Basin (Kimmig et al., 2021).

In the context of mantle-derived and fluids-dominated diagenetic settings, the hydrothermal dolomite phase is characterised by decreasing $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and increasing $\delta^{26}\text{Mg}$ values. This pattern explains the precipitation of hydrothermal dolomite in the upper Ediacaran Dengying Formation in the Nanshanping outcrop (Muñoz-López et al., 2024), the Devonian dolostones in the Rock Canyon Creek (Araoka et al., 2022), and the Permian dolostones of the Santanhu Basin (Jiao et al., 2023). These data shed light on the relative effects of crust- and mantle-derived fluids on

hydrothermal dolomitisation and offer a potential explanation for the formation mechanisms of ancient dolostones and their corresponding fluid sources.

Based on the analysis of the data published on various dolostones in Phanerozoic basins, hydrothermal dolomite is, for example, reported from Cambrian, Carboniferous, and Jurassic-Cretaceous systems (Fig. 1(b)). The Cambrian and Carboniferous time periods are characterised by large-scale igneous provinces (LIP) and silica LIP, respectively (Fig. 1(a)). Both LIP and silica LIP typify the Jurassic-Cretaceous. Notably, silicic LIP is a major crustal melting event and shows typical crustal characteristics compared to LIP (Bryan, 2007). These magnesium-rich deep-seated fluids migrate along faults and facilitate hydrothermal dolomitisation during these time intervals. Likewise, the multiple LIP peaks at 2.1, 1.4, and 0.8 Ga (Klausen, 2020) are in agreement with the Proterozoic hydrothermal dolomite abundance curve (Shang, 2023).

In summary, the multi-proxy approach utilising $\delta^{26}\text{Mg}$ and $\delta^{44}/^{40}\text{Ca}$ values as well as $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of phase-specific hydrothermal dolomite samples has significant potential to shed light on the sources of magnesium that triggered the dolomitisation process at depth. This includes contributions from mantle-derived fluids, crust-derived fluids (such as chlorite), heated seawater, and the mixing of different fluid types. Clearly, igneous provinces play a crucial role in the development of hydrothermal dolomite throughout geological time.

6. Conclusions

Using regionally important upper Ediacaran hydrothermal dolostone rock bodies in Sichuan Basin and Nanshanping outcrop in the Yangtze Block of China as a template, a multi-proxy geochemical and numerical approach is tested. Detailed analyses of $\delta^{26}\text{Mg}$, $\delta^{44}/^{40}\text{Ca}$, and $^{87}\text{Sr}/^{86}\text{Sr}$, along with the diagenetic modelling, resulted in a significantly improved understanding of the sources of Mg^{2+} in burial fluids. These are the main findings:

- Upper Ediacaran saddle dolomite in the upper Yangtze Block is characterised by comparably low $\delta^{26}\text{Mg}$ values ($-2.23 \pm 0.18\text{‰}$) and $\delta^{44}/^{40}\text{Ca}$ values (0.81‰ and 1.10‰), but high Sr concentrations (103.9 ± 56.0 ppm) and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (0.7119 ± 0.0017). This geochemical parameter set differs significantly from the host dolostone in the Sichuan Basin (Sr: 40 to 80 ppm; $^{87}\text{Sr}/^{86}\text{Sr}$: 0.7087 to 0.7094; $\delta^{26}\text{Mg}$: -2.2 to -1.8‰ ; $\delta^{44}/^{40}\text{Ca}$: ca. 1.2 ‰) and saddle dolomite sampled in the Nanshanping outcrop in the middle Yangtze Block (Sr: 47.0 ± 9.6 ppm; $\delta^{26}\text{Mg}$: -0.6‰ to -0.5‰). Hydrothermal dolomitisation in the burial domain of the Sichuan Basin is arguably related to Mg-bearing fluids that interacted with crustal rocks (chlorite) at depth, while the mixing between mantle-derived fluids and (heated) seawater likely led to the formation of saddle dolomite in the Nanshanping outcrop.
- Beyond the application of these tools to the case studies discussed here, the application of a combined $\delta^{26}\text{Mg}$, $\delta^{44}/^{40}\text{Ca}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ ratio geochemistry has proven instrumental to evaluate Mg^{2+} sources in burial brines in general. This is documented by applying these tools to a series of Precambrian and Phanerozoic case examples as documented in the literature. The dolomitising fluids at depth considered here include heated seawater, mantle- and crust-derived fluids, and various fluid mixing patterns. Geochemical data are validated and tested by diagenetic modelling, illustrating the potential geochemical trajectories from host dolostone (marine burial) to hydrothermal (deep burial) dolomite.

- (iii) Crust-derived and mantle-derived fluids contribute to the formation of mainly fault-related (fluid-buffered) replacement dolomite and dolomite (saddle) cement through geological time. This study is significant for those concerned with the occurrence and formation of hydrothermal dolomite and ancient dolomite.

CRediT authorship contribution statement

Yong-Jie Hu: Writing – review & editing, Writing – original draft, Project administration, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Chun-Fang Cai:** Writing – review & editing, Writing – original draft, Supervision, Funding acquisition, Conceptualization. **Ying Li:** Methodology, Investigation. **Lei Jiang:** Writing – review & editing. **Kai-Kai Li:** Writing – review & editing. **Shu-Yuan Shi:** Resources. **Zi-Wen Jiang:** Writing – review & editing. **Sylvia Riechermann:** Resources, Methodology. **Adrian Immenhauser:** Writing – review & editing, Writing – original draft.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This work has been financially supported by the Natural Science Foundation of China (42502117 and 42530805). Thanks to Kathrin Krimmler for sample preparation and analysis. Special thanks go to Editor Min Li for handling this manuscript and valuable comments. Four anonymous reviewers are likewise thanked for constructive improvements of the manuscript.

Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.petsci.2026.05.051>.

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